Light scattering from cold rolled aluminum surfaces

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Abstract

We present experimental light scattering measurements from aluminum surfaces obtained by cold rolling. We show that our results are consistent with a scale invariant description of the roughness of these surfaces. The roughness parameters that we obtain from the light scattering experiment are consistent with those obtained from atomic force microscopy measurements.

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Since an early paper by Berry in 1979 [1], the study of wave scattering from self-affine (fractal) surfaces has become very active, see Refs. [2–10] for recent references. Most of these papers consist in numerical simulations; apart from the early works of Jakeman [11] and Jordan et al. [12] very few theoretical results have been published; the same statement stands for experimental results while lots of real surfaces [13–15] have been shown to obey scale invariance. Here we try and test experimentally recent theoretical expressions obtained for the scattering of a scalar wave from a perfectly conducting self-affine surface [16]. We report scattering measurements of an s-polarized electromagnetic wave (632.8 nm) from a rough aluminum alloy plate (Al 5182). The latter was obtained by industrial cold rolling. As presented in Fig. 1 taken from Ref. [15] by Plouraboué and Boehm, the rolling process results in a very anisotropic surface, the roughness being much smaller along the rolling direction than in the orthogonal one. From atomic force microscopy (AFM) measurements with a long range scanner the authors could establish the scale invariant character of the roughness: the surface was found to be self-affine between a few tens of nanometers and about 50 μm. At the macroscopic scale, they measured the height standard deviation (RMS roughness) to be $\sigma = 2.5$ μm.

Let us briefly recall that a profile or a one-dimensional surface is said to be self-affine if it remains statistically invariant under the following transformations:

$$\Delta x \to \lambda x, \qquad \Delta z \to \lambda^{\zeta} z,$$

where the parameter ζ is the roughness exponent. A direct consequence of this scale invariance is

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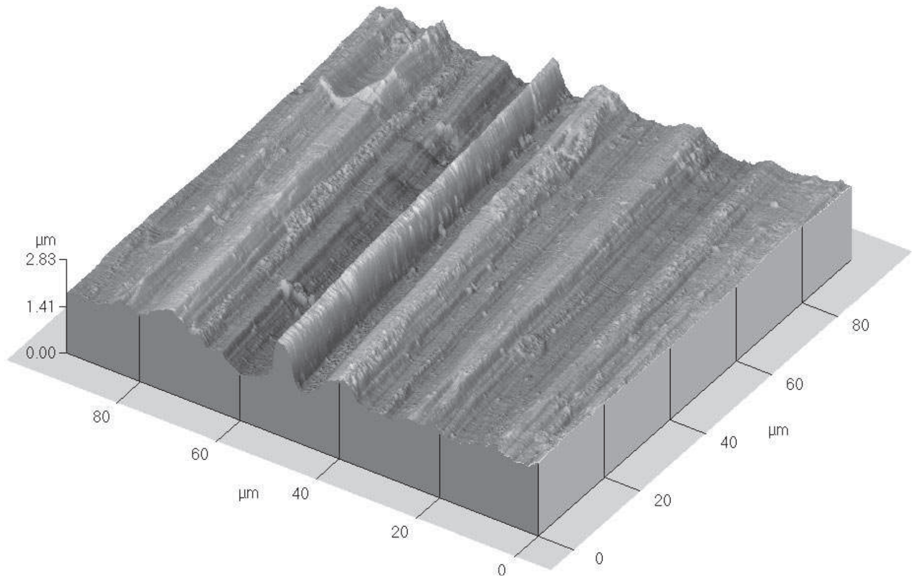

Fig. 1. AFM image of 512 × 512 points of the aluminum alloy sheet surface. This image has been obtained by Plouraboué and Boehm [15] in contact mode on a Park Scientific AFM using a long range scanner (100 µm lateral travel and 5 µm vertical travel). The height standard deviation has been measured to be $\sigma = 2.5$ µm.

that when measured over a length d geometrical quantities such as a roughness σ or a slope s are dependent on this length d:

$$\sigma(d) \propto d^{\zeta}, \qquad s(d) \propto d^{\zeta-1}.$$

The roughness exponent which characterizes the autocorrelation function is however not sufficient to give a complete characterization of the statistics of the surface roughness. The latter also requires an amplitude parameter. In the context of light scattering, one can for example normalize the geometrical quantities with their value over one wavelength:

$$\sigma(d) = \sigma(\lambda)\left(\frac{d}{\lambda}\right)^{\zeta}, \qquad s(d) = s(\lambda)\left(\frac{d}{\lambda}\right)^{\zeta-1}.$$

We will see in the following that the value of the slope $s(\lambda)$ is the crucial numerical parameter when dealing with scattering from self-affine rough surfaces. Note finally that the scale invariance of real surfaces roughness can only extend over a finite domain. The upper cut-off allows to define a macroscopic roughness, the lower one allows to define a local slope in every point. This scaling invariant formalism has been shown to be relevant to describe varied surfaces such as the ones obtained by fracture [13], growth or deposition processes [14].

We performed our measurements on a fully automated scatterometer (see Refs. [17,18] for a full description). The setup is designed for the measurement of the bidirectional scattering distribution function. The source is a helium–neon laser of wavelength $\lambda = 632.8$ nm, the beam passes through a mechanical chopper and is submitted to a spatial filtering before reaching the sample. The latter is placed on a rotating plate which allows to vary the incident angle. The scattered light is collected by a converging lens and focussed on a photomultiplier. This detection setup is placed on an automated rotating arm. Note that the shadow of the photomultiplier imposes a blind region of $\pm 11°$ around the back-scattering angle. Two polarizers allow us to select the polarization directions of both incident and scattered lights. The output signal is filtered by a lock-in amplifier and processed by a micro-computer. We used a frequency $f = 700$ Hz and a time constant $\tau = 1$ s. The surface being highly anisotropic, the result is a priori very sensitive to the orientation of the surface. In order to select properly one of the two main directions of the surface, we placed a vertical slit in front of the photomultiplier. This allows to reduce the effects of possible misorientation of the sample. The results of the scattering measurements obtained in s-polarization for incidence angles 0°, 30°, 50° and 65° are displayed in semi-log scale in Fig. 2.

How does the scale invariance of the roughness affect the angular distribution of the scattered light? The comparison of experimental light scattering data with theoretical models still remains a delicate matter. A key point is obviously to give a proper description of the statistical properties of the surface roughness. When testing new models or approximations, it is usual to design surfaces of controlled Gaussian autocorrelation function (this is for example possible by illuminating photosensitive materials with a series of laser speckles [19–21]). In the following we want to test the consistency of our scattering measurements with the roughness analysis. We perform this test via a very crude approximation: we consider the surface to be one dimensional and perfectly conducting. We then compare our experimental results with analytical predictions obtained in the context of a simple Kirchhoff approximation corresponding to Gaussian, exponential and self-affine correlations.

Although lots of studies have been published about scattering from scale invariant surfaces in the last 20 years, very few analytical results can be found in the literature. The main results are due to Jakeman and his collaborators [11,12] who showed that the angular distribution of the intensity of a wave scattered from a self-affine random phase screen could be written as a Lévy distribution. In a similar spirit, some of us studied very recently [16] the case of scattering of s-polarized waves from

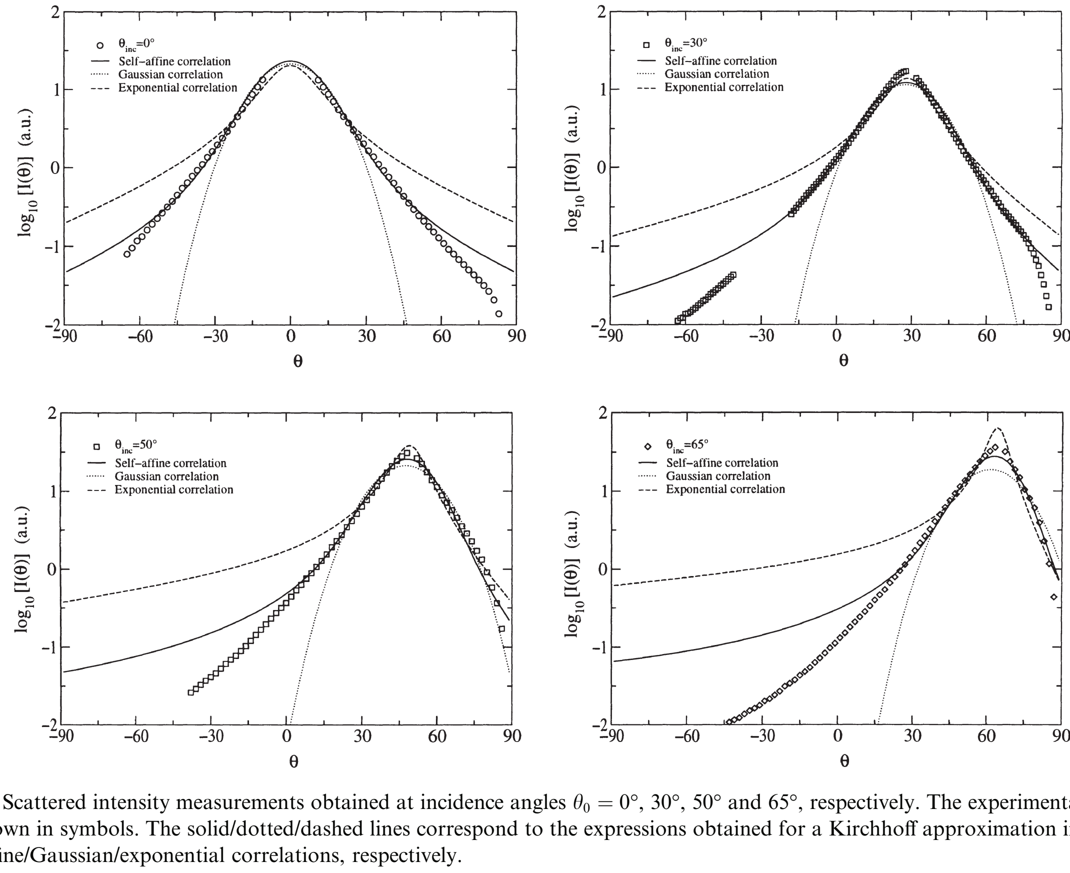

Fig. 2. Scattered intensity measurements obtained at incidence angles $\theta_0 = 0°$, 30°, 50° and 65°, respectively. The experimental results are shown in symbols. The solid/dotted/dashed lines correspond to the expressions obtained for a Kirchhoff approximation in case of self-affine/Gaussian/exponential correlations, respectively.

self-affine surfaces and found in the context of a Kirchhoff approximation the following expression for the scattering cross-section:

$$\left\langle \frac{\partial R_{\mathrm{s}}}{\partial \theta} \right\rangle = \frac{s(\lambda)^{-1/\zeta} a^{-((1/\zeta)-1)}}{\sqrt{2}\cos\theta_0} \times \frac{\cos\frac{\theta+\theta_0}{2}}{\cos^3\frac{\theta-\theta_0}{2}} \mathscr{L}_{2\zeta}\left(\frac{\sqrt{2}\tan\frac{\theta-\theta_0}{2}}{a^{(1/\zeta)-1} s(\lambda)^{1/\zeta}} \right), \quad (1)$$

where $a = 2\pi\sqrt{2}\cos((\theta+\theta_0)/2)\cos((\theta-\theta_0)/2)$, and $\mathscr{L}_\alpha(x)$ is the centered symmetrical Lévy stable distribution of exponent α defined as

$$\mathscr{L}_\alpha(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}k\, \mathrm{e}^{\mathrm{i}kx} \mathrm{e}^{-|k|^\alpha}. \quad (2)$$

Note that the form of this analytical result does not depend on the value of the global RMS roughness σ in contrast to the case of a Gaussian correlated surface. The scattering pattern is centered around the specular direction with an angular width w which scales as

$$w \simeq s(\lambda)^{1/\zeta}.$$

It is worth mentioning here that in the context of this simple Kirchhoff approximation, the crucial geometrical parameter to consider is the slope over the scale of one wavelength $s(\lambda)$: the angular distribution of the scattered intensity is mainly controlled by this "local" parameter and does not depend on the value of the global RMS roughness. The latter will only come back into the game if one goes beyond a single scattering approximation.

Using the complete set of experimental scattering data, we performed a numerical fitting procedure for the expression (1) and for the expressions obtained with Gaussian or exponential correlations. The latter have been derived in the case of very rough surfaces (see Appendix A for details of the expressions and the derivation). The fitting procedure consisted in a numerical minimization of the quadratic distance between the data and the tested expression in logarithmical scale. The free parameters are an amplitude parameter (which is simply an additive constant in logarithmic scale) and two geometrical parameters: the roughness exponent ζ and typical slope over the wavelength $s(\lambda)$. In the case of Gaussian or exponential correlation there is only one geometrical parameter which is an equivalent slope σ/τ or $2\pi\sigma^2/\lambda\tau$, respectively. Note that the same parameters are used for the whole set of experimental data gathering four different incidence angles.

In order to get rid of shadowing and multiple scattering effects, we restricted the fitting procedure to a region of $\pm 50°$ around the incidence angle. In this region we can see in Fig. 2 that there is a good agreement with the expression (1) which has been obtained with a roughness exponent $\zeta = 0.78$ and a typical slope over the wavelength $s(\lambda) = 0.11$. For large scattering angles the analytical expression systematically overestimates the scattered intensity. We attribute this behavior to the shadowing effects. None of the Gaussian and exponential correlations can give a comparable result. In the Gaussian case, we obtain $\sigma/\tau = 0.08$ and in the exponential case $2\pi\sigma^2/\lambda\tau = 0.10$.

Beyond this direct comparison of the different prediction for the angular distribution of the scattered intensity, we try also to compare the geometrical parameters that we obtained with direct roughness measurements performed by AFM. We imaged an area of size $2.048 \times 2.048\ \mu\mathrm{m}^2$ with a lateral step of 4 nm. From these roughness measurements we compute the typical height difference Δz between two points as a function of the distance Δx separating the two points. This quantity is obtained via a quadratic mean over all possible couples of points separated by a given distance Δx. In case of self-affine, Gaussian or exponential correlations, we expect respectively:

$$\Delta z_{\mathrm{sa}} = \lambda s(\lambda)\left(\frac{\Delta x}{\lambda}\right)^{\zeta}, \quad (3)$$

$$\Delta z_{\mathrm{Gauss}} = \sigma\sqrt{2}\sqrt{1-\exp\left(-\frac{\Delta x^2}{\tau^2}\right)}, \quad (4)$$

$$\Delta z_{\mathrm{exp}} = \sigma\sqrt{2}\sqrt{1-\exp\left(-\frac{\Delta x}{\tau}\right)}. \quad (5)$$

We show in Fig. 3 the results of the roughness analysis and the predictions corresponding to the self-affine correlations. Both the value $\zeta = 0.78$ of the roughness exponent and the slope over one wavelength $s(\lambda) = 0.11$ that we obtain from the

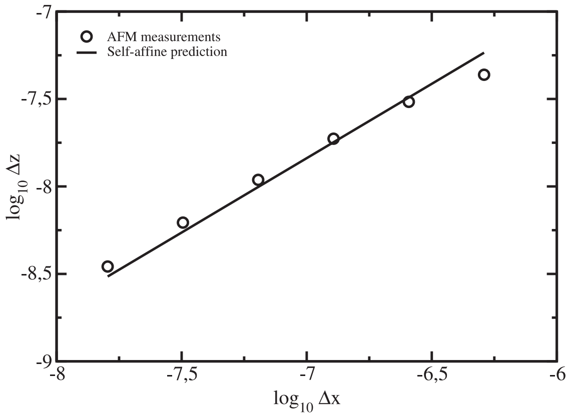

Fig. 3. Roughness analysis computed from AFM measurements (○) compared with predictions obtained via a fit of the angular scattered intensity distribution assuming self-affine correlations. The slope of the line is $\zeta = 0.78$ and the amplitude parameter is $s(\lambda) = 0.11$.

scattering measurements seem to be consistent with the experimental roughness data. Note that the hypothesis of exponential and Gaussian correlations would have lead to power laws of exponents 0.5 and 1, respectively, since we consider horizontal distances Δx about the wavelength which are far smaller than the expected correlation lengths.

These first results can be considered as very promising: let us recall that we assumed the surface to be purely one dimensional and perfectly conducting and that we used a basic Kirchhoff approximation, neglecting all shadowing or multiple scattering effects, etc. Refining the modeling of shadowing or multiple scattering in the specific case of self-affine surfaces could allow to design a valuable tool to measure the geometrical parameters describing self-affine surfaces. This experimental study also makes clear that self-affine correlations can be a relevant formalism to describe the optical properties of real surfaces. Beyond classical optical phenomena this could be also of great interest in the context of the recent studies [22,23] modeling thermal emission properties of rough surfaces.

Appendix A

We derive in this appendix the expression of the scattering cross-section in the framework of the Kirchhoff approximation for a one-dimensional very rough surface.

In the following we consider the scattering of s-polarized electromagnetic waves from a one-dimensional, rough surface $z = \zeta(x)$. The height distribution is supposed to be Gaussian of standard deviation σ and the two-points statistics is characterized by the autocorrelation function $C(v)$. The frequency of the wave is ω, the wave number is k, the incidence angle is θ_0, the scattering angle is θ.

Following Maradudin et al. [24] the Kirchhoff approximation gives for the scattering cross-section $\partial R_s / \partial \theta$ from a rough surface of infinite lateral extent:

$$\left\langle \frac{\partial R_s}{\partial \theta} \right\rangle = \frac{\omega}{2\pi c} \times \frac{1}{\cos\theta_0} \left(\frac{\cos[(\theta + \theta_0)/2]}{\cos[(\theta - \theta_0)/2]} \right)^2 \mathscr{I}(\theta, \theta_0), \tag{A.1}$$

where

$$\mathscr{I}(\theta, \theta_0) = \int_{-\infty}^{\infty} \mathrm{d}v \exp\{\mathrm{i}k(\sin\theta - \sin\theta_0)v\} \Omega(v), \tag{A.2}$$

$$\Omega(v) = \langle \exp\{-\mathrm{i}k[\cos\theta + \cos\theta_0]\Delta\zeta(v)\}\rangle. \tag{A.3}$$

Note that the statistical properties of the profile function, $\zeta(x)$, enters Eq. (A.1) only through $\Omega(v)$. With the knowledge of the autocorrelation function $C(v)$ the distribution of the height differences $\Delta\zeta(v) = \zeta(x+v) - \zeta(x)$ can be written:

$$P(\Delta\zeta, v) = \frac{1}{2\sigma\sqrt{\pi}\sqrt{1 - C(v)}} \exp\left[\frac{-\Delta\zeta^2}{4\sigma^2[1 - C(v)]}\right]. \tag{A.4}$$

This leads immediately to:

$$\Omega(v) = \exp\{-k^2\sigma^2(\cos\theta + \cos\theta_0)^2[1 - C(v)]\}. \tag{A.5}$$

In case of a very rough surface, we have $k^2\sigma^2 \ll 1$ (in our experimental case, $\sigma = 2.5$ μm and $\lambda = 632.8$ nm so that $k^2\sigma^2 \simeq 600$) and the only v to really contribute to the integral are in the close vicinity of zero. We can then replace $C(v)$ by the first terms of its expansion around zero. Consider the Gaussian and exponential cases

$$C_{\mathrm{G}}(v) = \exp\left(-\frac{v^2}{\tau^2}\right), \qquad C_{\exp}(v) = \exp\left(-\frac{v}{\tau}\right), \tag{A.6}$$

where τ is by definition the correlation length, this leads to:

$$\Omega_{\mathrm{G}}(v) = \exp\left[-k^2(\cos\theta + \cos\theta_0)^2\alpha^2 v^2\right], \tag{A.7}$$

$$\Omega_{\exp}(v) = \exp\left[-k^2(\cos\theta + \cos\theta_0)^2\alpha\sigma|v|\right]. \tag{A.8}$$

Simple algebra leads finally to

$$\left\langle \frac{\partial R_{\mathrm{s}}}{\partial\theta} \right\rangle_{\mathrm{G}} = \frac{k}{4\alpha\sqrt{\pi}\cos\theta_0}\frac{\cos[(\theta+\theta_0)/2]}{\cos^3[(\theta-\theta_0)/2]} \times \exp\left[-\frac{1}{4\alpha^2}\left(\tan\frac{\theta-\theta_0}{2}\right)^2\right], \tag{A.9}$$

$$\left\langle \frac{\partial R_{\mathrm{s}}}{\partial\theta} \right\rangle_{\exp} = (\alpha\sigma k/\pi\cos\theta_0)(\cos^2[(\theta+\theta_0)/2] / \{\sin^2[(\theta-\theta_0)/2] + 4(\alpha\sigma k)^2\cos^2[(\theta+\theta_0)/2] \times \cos^4[(\theta-\theta_0)/2]\}). \tag{A.10}$$

References

[1] M.V. Berry, J. Phys. A 12 (1979) 781.
[2] D.L. Jaggard, X. Sun, J. Appl. Phys. 68 (11) (1990) 5456.
[3] M.K. Shepard, R.A. Brackett, R.E. Arvidson, J. Geophys. Res. 100 (E6) (1995) 11709.
[4] P.E. McSharry, P.J. Cullen, D. Moroney, J. Appl. Phys. 78 (12) (1995) 6940.
[5] N. Lin, H.P. Lee, S.P. Lim, K.S. Lee, J. Mod. Opt. 42 (1995) 225.
[6] J. Chen, T.K.Y. Lo, H. Leung, J. Litva, IEEE Trans. Geosci. 34 (4) (1996) 966.
[7] C.J.R. Sheppard, Opt. Commun. 122 (1996) 178.
[8] J.A. Sánchez-Gil, J.V. García-Ramos, Waves Random Media 7 (1997) 285.
[9] J.A. Sánchez-Gil, J.V. García-Ramos, J. Chem. Phys. 108 (1) (1998) 317.
[10] Y.-P. Zhao, C.F. Cheng, G.C. Wang, T.M. Lu, Surf. Sci. 409 (1998) L703.
[11] E. Jakeman, in: L. Pietronero, E. Tossati (Eds.), Fractals in Physics, Elsevier, Amsterdam, 1986.
[12] D.L. Jordan, R.C. Hollins, E. Jakeman, A. Prewett, Surf. Topography 1 (1988) 27.
[13] E. Bouchaud, J. Phys., Condens. Matt. 9 (1997) 4319.
[14] P. Meakin, Fractals, Scaling and Growth Far from Equilibrium, Cambridge University Press, Cambridge, 1998.
[15] F. Plouraboué, M. Boehm, Trib. Int. 32 (1999) 45.
[16] I. Simonsen, D. Vandembroucq, S. Roux, Phys. Rev. E 61 (2000) 5914.
[17] J.J. Greffet, Ph.D. Thesis, Université Paris Sud, Orsay, France, 1988.
[18] J.J. Greffet, in: R.P. Breault (Ed.), Stray Light and Contamination in Optical Systems, Proc. Soc. Photo-Opt. Instrum. Eng. 967 (1989) 184.
[19] M.E. Knotts, T.R. Michel, K.A. O'Donnel, J. Opt. Soc. Am. A 10 (1993) 928.
[20] M.E. Knotts, K.A. O'Donnel, J. Opt. Soc. Am. A 11 (1994) 697.
[21] O. Calvo-Perez, Ph.D. Thesis, Ecole Centrale Paris,1998.
[22] R. Carminati, J.J. Greffet, Phys. Rev. Lett. 82 (8) (1999) 1660.
[23] J.-J. Greffet, R. Carminati, Heat & Technol. 18 (2000) 81.
[24] A.A. Maradudin, T. Michel, A.R. McGurn, E.R. Méndez, Ann. Phys. 203 (1990) 255.