

On the role of flexoelectricity in triboelectricity for randomly rough surfaces

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Abstract – I show how flexoelectricity results in a fluctuating surface electric potential when elastic solids with random roughness are squeezed into contact. The flexoelectric potential may induce surface charge distributions and hence contribute to triboelectricity. Using the developed theory I analyze the Kelvin Force Microscopy data of Baytekin *et al.* for the electric potential above a polydimethylsiloxane (PDMS) surface after it was peeled away from another PDMS surface.

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Introduction. – When two solids in adhesive contact are pulled apart they are often left charged. More generally, fluctuating charge distributions (both positive and negative charge distributions on both solids) are left on the surfaces after sliding contact or pull-off [1]. This triboelectric effect has been known for more than 25 centuries and has important practical implications. However, the origin of the charge separation is not well understood [2].

In a recent paper Mizzi *et al.* [3] have proposed that triboelectricity often results from flexoelectricity. The flexoelectric effect consists of the linear coupling between strain gradient and electric polarization [4,5]. When two solids are squeezed into contact very large strain gradients will occur in the asperity contact regions, and Mizzi *et al.* have shown that this may result in large flexoelectric potential differences at the nanoscale. If free charges exist (in the solids or in the atmosphere) they may move or rearrange in such a way as to screen the flexoelectric potential field, and one may end up with charge distributions at, or close to, the solid surfaces which follow the spatial variation of the flexoelectric potential field.

Mizzi *et al.* studied how the flexoelectric surface potential depends on the normal force when a sphere is squeezed against an elastic half-space. For this they used the well-known results for the strain at Hertz (or JKR) sphere-flat type of contact [6]. However, the contact between two randomly rough surfaces cannot be described by a model assuming independent Hertzian contact regions since the long-ranged elastic coupling between the contact regions strongly affects the nature of the contact [7,8]. In this paper I show how the flexoelectric potential can be calculated

for the contact between elastic solids with randomly rough surfaces.

Theory. – Mizzi *et al.* (see ref. [3]) have shown that the normal component of the electric field induced by a flexoelectric coupling in an isotropic nonpiezoelectric half-space is given (approximately) by

$$E_z = -f(\epsilon_{ii,z} + 2\epsilon_{zi,i}), \quad (1)$$

where $\epsilon_{ij,k} = \partial\epsilon_{ij}/\partial x_k$, where

$$\epsilon_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) \quad (2)$$

is the strain tensor. Here u_i are the components of the displacement field $\mathbf{u}(\mathbf{x}, t)$. In (1) summation over repeated indices is implicitly understood. The flexocoupling voltage f can be both positive or negative, and for polymers $|f|$ is typically in the range 5–300 V (see table S2 in ref. [3]). Substituting (2) in (1) gives

$$E_z = -f \left(\nabla^2 u_z + 2 \frac{\partial}{\partial z} \nabla \cdot \mathbf{u} \right). \quad (3)$$

Assuming a solid with homogeneous and isotropic elastic properties, from the theory of elasticity the displacement field \mathbf{u} satisfies

$$\rho \frac{\partial^2 \mathbf{u}}{\partial t^2} = \mu \nabla^2 \mathbf{u} + (\mu + \lambda) \nabla \nabla \cdot \mathbf{u}, \quad (4)$$

where ρ is the mass density, and where the Lamé constants λ and μ can be related to the Young's elastic modulus E

and the Poisson ratio ν via

$$\lambda = \frac{\nu E}{(1+\nu)(1-2\nu)}, \quad \mu = \frac{E}{2(1+\nu)}. \quad (5)$$

In principle the equation determining the deformation field \mathbf{u} should be influenced by the flexoelectric field, and the charge rearrangement which may occur as a result of it, but this effect will be assumed small in what follows. We neglect the time dependency so that from (4)

$$\nabla^2 \mathbf{u} = - \left(1 + \frac{\lambda}{\mu}\right) \nabla \nabla \cdot \mathbf{u}. \quad (6)$$

Using (3) and (6) gives

$$E_z = - \left(1 - \frac{\lambda}{\mu}\right) f \frac{\partial}{\partial z} \nabla \cdot \mathbf{u}. \quad (7)$$

The electric potential ϕ at the surface ($z = 0$) relative to far inside the solid ($z = \infty$) is equal to

$$\phi = - \int_0^\infty dz E_z = - \left(1 - \frac{\lambda}{\mu}\right) f \nabla \cdot \mathbf{u}, \quad (8)$$

where $\nabla \cdot \mathbf{u}$ is evaluated for $z = 0$.

For randomly rough surfaces the electric potential $\phi(\mathbf{x})$ will vary in a complex way with the coordinate $\mathbf{x} = (x, y)$ on the surface $z = 0$. Here we consider first the mean square of the (fluctuating) electric potential, $\langle \phi^2(\mathbf{x}) \rangle$, and then the electric potential power spectrum, which contains more information about the fluctuating electric potential.

Mean square fluctuation of the electric potential. Consider $\langle \phi^2 \rangle$, where $\langle \cdot \rangle$ stands for ensemble average. The ensemble average of $\phi^2(\mathbf{x})$ is assumed to be independent of \mathbf{x} so that

$$\langle \phi^2 \rangle = \frac{1}{A_0} \int d^2x \langle \phi^2(\mathbf{x}) \rangle, \quad (9)$$

where A_0 is the surface area. We write

$$\phi(\mathbf{x}) = \int d^2q \phi(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{x}}. \quad (10)$$

From (9) and (10) we get

$$\langle \phi^2 \rangle = \frac{(2\pi)^2}{A_0} \int d^2q \langle \phi(-\mathbf{q}) \phi(\mathbf{q}) \rangle. \quad (11)$$

We can calculate $\nabla \cdot \mathbf{u}$ using the formalism presented in appendix A in ref. [9]. Assume that a stress $\sigma_i(\mathbf{x}, t)$ acts on the surface of an elastic half-space. We write

$$\sigma_i(\mathbf{x}, t) = \int d^2q d\omega \sigma_i(\mathbf{q}, \omega) e^{i(\mathbf{q} \cdot \mathbf{x} - \omega t)}. \quad (12)$$

We note here that although we have assumed above that the time dependency of the flexoelectric field can be neglected, it is in the present approach necessary to include it in the calculation of $\nabla \cdot \mathbf{u}$, and only at the end let $\omega \rightarrow 0$ corresponding to a time-independent problem.

The displacement field in the solid for $z > 0$ is written as (see appendix A in ref. [9])

$$\mathbf{u} = \mathbf{p}A + \mathbf{K}B + \mathbf{p} \times \mathbf{K}C, \quad (13)$$

where $\mathbf{p} = -i\nabla$ and where $\mathbf{K} = \mathbf{n} \times \mathbf{p}$, where \mathbf{n} is a unit vector normal to the surface pointing along the z -axis. Thus we get

$$\nabla \cdot \mathbf{u} = i\mathbf{p} \cdot \mathbf{u} = ip^2 A = -i\nabla^2 A. \quad (14)$$

The scalar potential A satisfies the wave equation (see (A4) in ref. [9]), which with the Fourier-transformed time variable takes the form

$$\nabla^2 A + \frac{\omega^2}{c_L^2} A = 0.$$

Using this in (14) we get

$$\nabla \cdot \mathbf{u} = i \frac{\omega^2}{c_L^2} A. \quad (15)$$

Using (A19) in ref. [9] we get for $z = 0$, with the Fourier-transformed \mathbf{x} and t dependency,

$$i \frac{\omega^2}{c_L^2} A = \frac{\omega^2}{c_L^2} \frac{1}{\mu S} \left[2p_T \mathbf{q} + \left(\frac{\omega^2}{c_T^2} - 2q^2 \right) \mathbf{n} \right] \cdot \sigma, \quad (16)$$

where

$$S = \left(\frac{\omega^2}{c_T^2} - 2q^2 \right)^2 + 4q^2 p_T p_L, \quad (17)$$

where

$$p_T = \left(\frac{\omega^2}{c_T^2} - q^2 + i0^+ \right)^{1/2}, \quad p_L = \left(\frac{\omega^2}{c_L^2} - q^2 + i0^+ \right)^{1/2},$$

where 0^+ is an infinitesimal small positive number and where c_T and c_L are the transverse and longitudinal sound velocities, respectively. Using (16) and (17) for $\omega \rightarrow 0$ we get

$$i \frac{\omega^2}{c_L^2} A = \frac{1}{\lambda + \mu} (-i\hat{q} + \mathbf{n}) \cdot \sigma, \quad (18)$$

where $\hat{q} = \mathbf{q}/q$. If we denote $\mathbf{e} = -i\hat{q} + \mathbf{n}$ we get from (8), (11), (15) and (18)

$$\langle \phi^2 \rangle = \left(\frac{\kappa f}{E} \right)^2 \frac{(2\pi)^2}{A_0} \int d^2q e_i^* e_j \langle \sigma_i(-\mathbf{q}) \sigma_j(\mathbf{q}) \rangle, \quad (19)$$

where

$$\kappa = \frac{(\lambda - \mu)E}{\mu(\mu + \lambda)} = 2(4\nu - 1)(1 + \nu). \quad (20)$$

For rubber-like materials $\nu \approx 0.5$ giving $\kappa \approx 3$.

We now consider the simplest case of pull-off without sliding. In this case the stress will be approximately normal to the surface and (19) reduces to

$$\langle \phi^2 \rangle = \left(\frac{\kappa f}{E} \right)^2 \frac{(2\pi)^2}{A_0} \int d^2q \langle \sigma(-\mathbf{q}) \sigma(\mathbf{q}) \rangle, \quad (21)$$

where $\sigma(\mathbf{q})$ now denotes the normal stress component. Now, since

$$\int d^2q \langle \sigma(-\mathbf{q})\sigma(\mathbf{q}) \rangle = \frac{A_0}{(2\pi)^2} \langle \sigma^2(\mathbf{x}) \rangle, \quad (22)$$

we can write

$$\langle \phi^2 \rangle = \left(\frac{\kappa f}{E} \right)^2 \langle \sigma^2 \rangle. \quad (23)$$

If $P(\sigma, p_0)$ is the probability distribution of stresses at the interface, which depends on the applied stress p_0 , then

$$\langle \sigma^2 \rangle = \int_{-\infty}^{\infty} d\sigma \sigma^2 P(\sigma, p_0). \quad (24)$$

The probability distribution $P(\sigma, p_0)$ can be calculated for randomly rough surfaces, both with and without the adhesion, using the Persson contact mechanics theory [10], or using numerically (exact) methods such as the boundary element method [8,11]. Here we consider first the non-adhesive contact between an elastic half-space and a rigid countersurface, where the roughness is characterized by the surface roughness power spectrum $C_{hh}(q)$. For non-adhesive contact $P(\sigma, p_0) = 0$ for $\sigma < 0$ and for $\sigma > 0$ (see ref. [12]):

$$P = \frac{1}{s(2\pi)^{1/2}} \left[e^{-(\sigma-p_0)^2/(2s^2)} - e^{-(\sigma+p_0)^2/(2s^2)} \right], \quad (25)$$

where $s = E^*h'/2$, where $E^* = E/(1-\nu^2)$ and $h' = \langle (\nabla h)^2 \rangle^{1/2}$ is the rms slope of the rough surface with the surface profile $z = h(\mathbf{x})$. For $p_0/E^* \ll 1$ the probability distribution reduces to

$$P \approx \left(\frac{2}{\pi} \right)^{1/2} \frac{\sigma p_0}{s^3} \exp\left(-\frac{\sigma^2}{2s^2}\right). \quad (26)$$

In this limit we get from (24) and (26)

$$\langle \sigma^2 \rangle = 2 \left(\frac{2}{\pi} \right)^{1/2} s p_0, \quad (27)$$

so that

$$\langle \phi^2 \rangle = \left(\frac{\kappa f}{E} \right)^2 2 \left(\frac{2}{\pi} \right)^{1/2} s p_0 \quad (28)$$

or

$$\langle \phi^2 \rangle = \left[\left(\frac{32}{\pi} \right)^{1/2} \frac{1+\nu}{1-\nu} (1-4\nu)^2 \right] f^2 h' \frac{p_0}{E}. \quad (29)$$

In the limit when $A/A_0 \ll 1$, and when the surface roughness power spectrum has a wide roll-off region, the contact regions will consist of a low concentration of small contact patches as indicated in fig. 1. If the number of contact patches is denoted by N and if the average area of a contact patch is A_1 then $A = NA_1$. If the mean square value of the voltage at the surface of a contact region is ϕ_1^2 then

$$\langle \phi^2 \rangle A_0 = N \phi_1^2 A_1$$

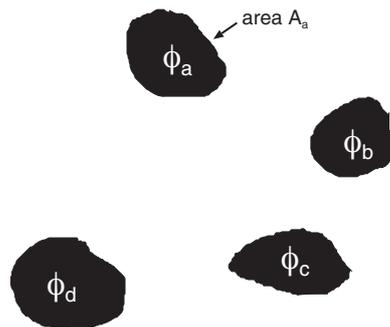


Fig. 1: Contact patches between two elastic solids with random roughness. The electric potential in the contact regions fluctuates between positive and negative values. The average contact patch area is denoted by A_1 and the rms electric potential in a contact patch by ϕ_1 .

or

$$\phi_1^2 = \langle \phi^2 \rangle \frac{A_0}{A}. \quad (30)$$

From (26) we get the relative contact area (valid for $A/A_0 \ll 1$) (see also ref. [10])

$$\frac{A}{A_0} = \int_0^{\infty} d\sigma P \approx \frac{p_0}{s} = \left(\frac{8}{\pi} \right)^{1/2} \frac{p_0}{h'E^*}. \quad (31)$$

From (29)–(31) we get

$$\phi_1 \approx \xi |f| h', \quad (32)$$

where

$$\xi = \sqrt{2} \frac{|1-4\nu|}{1-\nu}. \quad (33)$$

The derivation of (32) is strictly valid only if all the contact spots have the same area. In reality, this is not the case, and there will be a (probability) distribution P_A of contact sizes. In fact, for self-affine fractal surfaces numerical studies have shown a power-law size distribution (see, *e.g.*, ref. [13,14]). However, in this case (32) is still valid if we define

$$\phi_1^2 = \frac{\int dA P_A A \bar{\phi}^2(A)}{\int dA P_A A},$$

where $\bar{\phi}^2(A)$ is the mean square voltage in a contact spot with the area A , where the average (indicated by the overbar) is over the shape of all contact regions with the area A .

Another interesting limiting case is contact with adhesion when the adhesion is so strong as to pull the solids into complete contact. This limit is easy to study: First note that complete contact prevails as $p_0 \rightarrow \infty$ and in this limit we can neglect the second term in (25). This gives a Gaussian-like probability distribution centered at $\sigma = p_0$. However, with adhesion we consider the case without an applied pressure *i.e.*, $p_0 = 0$. Thus for adhesion and assuming complete contact with $p_0 = 0$ we have the (exact)

stress probability distribution

$$P = \frac{1}{s(2\pi)^{1/2}} e^{-\sigma^2/(2s^2)}, \quad (34)$$

which is valid for all σ (both positive and negative). Using (34) we get

$$\langle \sigma^2 \rangle = \frac{1}{s(2\pi)^{1/2}} \int_{-\infty}^{\infty} d\sigma \sigma^2 e^{-\sigma^2/(2s^2)} = s^2. \quad (35)$$

Substituting (35) in (23) gives

$$\langle \phi^2 \rangle = \left(\frac{\kappa f h'}{2(1-\nu^2)} \right)^2 \quad (36)$$

or

$$\langle \phi^2 \rangle^{1/2} = \xi' |f| h', \quad (37)$$

where

$$\xi' = \frac{4\nu - 1}{1 - \nu} \quad (38)$$

For rubber-like materials $\nu \approx 0.5$ giving $\xi \approx 2.8$ and $\xi' \approx 2.0$. As an example, for natural rubber $|f| \approx 20$ V and if we assume the rms slope $h' \approx 0.1$ we get the average electric potential in the asperity contact regions $\phi_1 \approx 5$ V, and for complete contact the rms surface electric potential $\langle \phi^2 \rangle^{1/2} \approx 4$ V.

Electric potential power spectrum. Let us now study the electric potential power spectrum

$$C_{\phi\phi}(\mathbf{q}) = \frac{(2\pi)^2}{A_0} \langle |\phi(\mathbf{q})|^2 \rangle. \quad (39)$$

We can calculate $C_{\phi\phi}$ from the theory presented above. Thus from (8), (15) and (18)

$$\phi(\mathbf{q}) = - \left(1 - \frac{\lambda}{\mu} \right) f \frac{1}{\lambda + \mu} \mathbf{e} \cdot \sigma. \quad (40)$$

Thus we get

$$C_{\phi\phi}(\mathbf{q}) = \frac{(2\pi)^2}{A_0} \left(\frac{\mu - \lambda}{\mu(\mu + \lambda)} \right)^2 f^2 e_i^* e_j \langle \sigma_i(-\mathbf{q}) \sigma_j(\mathbf{q}) \rangle. \quad (41)$$

If we assume no shear forces we get

$$C_{\phi\phi}(\mathbf{q}) = \frac{(2\pi)^2}{A_0} \left(\frac{\mu - \lambda}{\mu(\mu + \lambda)} \right)^2 f^2 \langle \sigma(-\mathbf{q}) \sigma(\mathbf{q}) \rangle, \quad (42)$$

where $\sigma(\mathbf{q})$ is the normal stress. In ref. [7] we have shown that to a good approximation

$$\langle \sigma(-\mathbf{q}) \sigma(\mathbf{q}) \rangle \approx \left(\frac{\mu}{1 - \nu} \right)^2 \frac{A_0}{(2\pi)^2} q^2 C_{hh}(q) P(q). \quad (43)$$

Here $C_{hh}(q)$ is the surface roughness power spectrum,

$$C_{hh}(q) = \frac{1}{(2\pi)^2} \int d^2x \langle h(\mathbf{x}) h(\mathbf{0}) \rangle e^{-i\mathbf{q} \cdot \mathbf{x}},$$

where $z = h(\mathbf{x})$ is the surface height coordinate. In (43) $P(q) = A(q)/A_0$ is the relative contact area when only the roughness components with the wavenumber smaller than q is included when calculating $A(q)$ (see ref. [10]).

Substituting (43) in (42) gives

$$C_{\phi\phi} \approx \left(\frac{\mu - \lambda}{\mu + \lambda} \right)^2 \left(\frac{f}{1 - \nu} \right)^2 q^2 C_{hh}(q) P(q). \quad (44)$$

For rubber materials $\nu \approx 0.5$ and $\lambda \approx \infty$ giving

$$C_{\phi\phi} \approx 4f^2 q^2 C_{hh}(q) P(q). \quad (45)$$

If we assume complete contact between the solids at the interface then $P(q) = 1$ and

$$C_{\phi\phi} \approx 4f^2 q^2 C_{hh}(q). \quad (46)$$

Many surfaces are self-affine fractals with a roll-off region for $q < q_0$. For such surfaces [15]:

$$C_{hh} = C_0, \quad \text{for } q < q_0, \quad (47)$$

$$C_{hh} = C_0 \left(\frac{q}{q_0} \right)^{-2(1+H)}, \quad \text{for } q > q_0, \quad (48)$$

where

$$C_0 = \frac{1}{\pi} \frac{H}{1 + H} \frac{\langle h^2 \rangle}{q_0^2}, \quad (49)$$

where $\langle h^2 \rangle$ is the mean square roughness and where the Hurst exponent H is between 0 and 1 but typically $H \approx 1$ (see ref. [15]). The latter is expected for sandblasted surfaces and, at least in some cases, for contaminated surfaces [16], which can be considered as generated by a process opposite to sandblasting (depositing of particles rather than removal of particles). Substituting (47) and (48) in (46) gives an electric potential power spectrum which scales $\sim q^{-2H}$ for $q > q_0$ and q^2 for $q < q_0$. For $q = q_0$

$$C_{\phi\phi} \approx 4f^2 q_0^2 C_0, \quad (50)$$

which gives

$$\langle h^2 \rangle = \frac{\pi}{4} \frac{1 + H}{H} f^{-2} C_{\phi\phi}, \quad (51)$$

where $C_{\phi\phi}$ is evaluated for $q = q_0$.

Discussion. – The flexoelectric effect gives rise to an electric field in the surface region between contacting solids. This field will drive charges (either from the gas phase or from the solids) to rearrange in such a way as to screen out the electric field as completely as possible. Following Mizzi *et al.* we will assume that when the solid bodies are separated this will result in a surface distribution of charges which will generate an electric potential outside of the solids. The charge distribution will of course decay with increasing time but a short time after

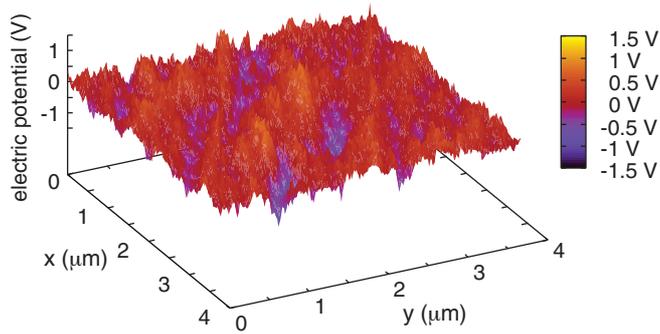


Fig. 2: The measured electric potential a distance ≈ 100 nm above a PDMS surface [1].

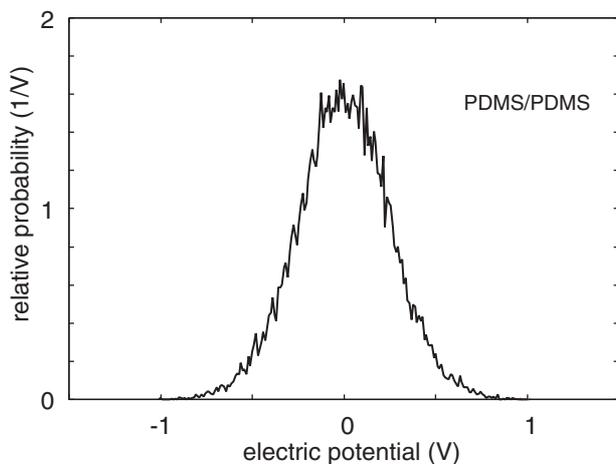


Fig. 3: The probability distribution of the electric potential $P(\phi)$ a distance $d \approx 100$ nm above a PDMS surface. The rms voltage fluctuation from the measured data is $\langle \phi^2 \rangle^{1/2} \approx 0.26$ V.

the surface separation it may result in an electric potential distribution of similar form as that generated by the flexoelectric effect. This electric potential can be studied using Kelvin Force Microscopy.

In ref. [1] Baytekin *et al.* have used Kelvin Force Microscopy in a study of the electric potential $\phi(\mathbf{x})$ at a distance ≈ 100 nm above a PDMS surface, a short time after separating it from the contact with another PDMS surface. The PDMS surfaces were prepared by molding the rubber against an atomically flat (silanized) silicone wafer. However, the silicone wafer was exposed to the normal atmosphere and may have a contamination film so the PDMS sheets may have surface roughness, with a rms roughness amplitude of the order of a few nanometer.

Because of the small surface roughness the two PDMS sheets are likely to be in complete adhesive contact. After separation the surfaces have charge distributions which oscillate between positive and negative values (see fig. 2) such that the net charge is small compared to the total number of charges. This is clear from the probability distribution $P(\phi)$ of the electric potential shown in fig. 3. Note that $P(\phi)$ is nearly a perfect Gaussian centered at $\phi \approx 0$ *i.e.*, the net charge is very small.

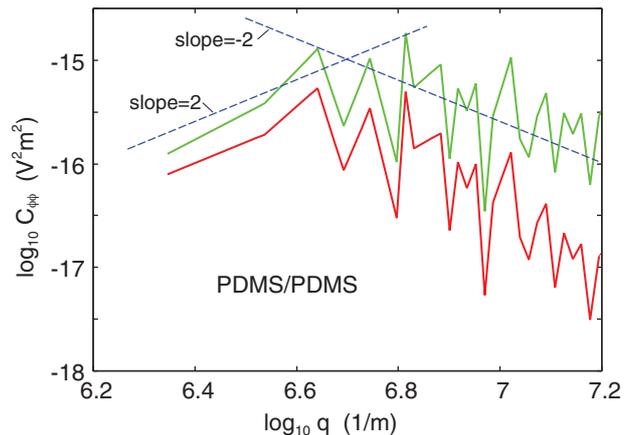


Fig. 4: The electric potential power spectrum $C_{\phi\phi}$ as a function of the wavenumber q (log-log scale). The red line is the power spectrum using the measured electric potential which was obtained with the Kelvin Force Microscopy tip ≈ 100 nm above the PDMS surface. The green line is the power spectrum at the PDMS surface assuming that the power spectrum depends on the tip-substrate separation d as $\exp(-2qd)$ (see ref. [17]). The blue dashed lines have the slope +2 and -2.

Figure 4 shows the electric potential power spectrum. The data is very noisy due to the rather small number of Kelvin Force Microscopy data points (152×152). We do not show results for $q > 1.6 \times 10^7 \text{ m}^{-1}$ (or $\log_{10} q > 7.2$) because $C_{\phi\phi}$ for large q is influenced by the fact that the scanning tip was located ≈ 100 nm above the PDMS surface. The red line in fig. 4 is obtained directly from the measured electric potential, and the green line is the power spectrum at the PDMS surface, assuming the power spectrum decays like $\exp(-2qd)$ with the separation d of the probe tip from the PDMS surface [17]. Note that for small wavenumber $q < q_0 \approx 4 \times 10^6 \text{ m}^{-1}$ the power spectrum increases like q^2 with the wavenumber, while for $q > q_0$ it decreases roughly as q^{-2} ; both results are expected from the theory above if the Hurst exponent $H \approx 1$ and if a flat roll-off region occurs in the roughness power spectrum for $q < q_0$. Baytekin *et al.* also performed studies for PDMS pulled off from a smooth polycarbonate (PC) surface, but the results are very similar to those for PDMS against PDMS. We do note however that the region for $q < q_0$ in fig. 4 is very uncertain due to the small number of long-wavelength roughness components. Thus, a more accurate study requires Kelvin Force Microscopy measurements over a larger surface area.

We can use (51) to estimate the rms roughness necessary in order to reproduce the magnitude of the observed electric potential power spectrum. Thus for $q = q_0$ from fig. 4 we get $C_{\phi\phi} \approx 8 \times 10^{-16} \text{ V}^2\text{m}^2$ and using (51) with $H = 1$ this gives $\langle h^2 \rangle^{1/2} \approx 6 \text{ nm}$ if $f = 6 \text{ V}$. This value is very reasonable for a surface where the roughness is produced by a contamination film due to the exposure of the wafer to the normal atmosphere. The roll-off wavelength $\lambda_0 = 2\pi/q_0 \approx 1 \mu\text{m}$ also appears very reasonable.

Summary and conclusion. – We have presented a theory for the electric potential at a surface produced by flexoelectricity for elastic solids with randomly rough surfaces. We have calculated the power spectrum of the electric potential $\phi(\mathbf{x})$. In the light of the theory we have discussed the experimental contact electrification results of Baytekin *et al.* and found good correlation with the theory predictions.

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