

Surface roughness of peeled adhesive tape: A mystery?

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Abstract – We study the surface roughness of adhesive tape peeled in stick-slip. We find two regions: a smooth surface region associated with fast crack propagation and a rough surface region associated with slow crack propagation. In both regions the surfaces may be self-affine fractal-like with the fractal dimension $D_f \approx 2.3$. This fractal dimension is typical for surfaces produced by crack propagation, but unexpected in the present case.

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Surfaces produced by crack propagation are known to often exhibit self-affine fractal scaling properties with the fractal dimension $D_f \approx 2.2-2.4$ (see refs. [1–4]). Here we show that this may be true even for elastically very soft materials, namely the tacky rubber layer on adhesive tapes, when pulled from a roll. However, as we will argue below, this result is unexpected because of the smoothing influence of the surface tension.

Two different modes of peeling adhesive tape have been observed involving steady and non-steady crack-tip motion. Steady peeling at low velocity result in a rubber surface which appears white due to light scattering from relative large surface roughness. Steady peeling at high velocity result in a much smoother (and transparent) rubber film. However, as a function of the crack-tip velocity there is a gradual change in the surface topography. At low crack-tip velocity small detached regions at the adhesive-substrate interface (see fig. 1(a)) (which may occur naturally, *e.g.*, as a result of interfacial roughness, or which may nucleate due to the high tensile stress in front of the peeling edge) will expand into the soft rubber which may result in long polymer filaments [5–7], which finally break (via interfacial crack propagation) at the rubber-substrate interface. During the resulting fast snapping motion the elastic energy stored in the elongated filament (long bond) is dissipated, which will result in a very large crack propagation energy $G(v)$ at low peeling velocity. As the peeling velocity increases, the effective elastic modulus of the rubber increases, which result in less displacement of the detached area into the rubber film,

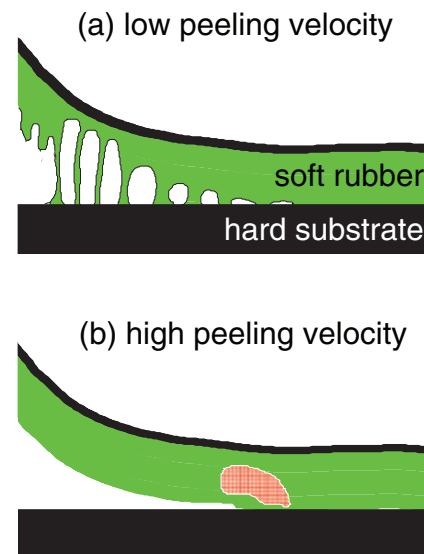


Fig. 1: (Colour on-line) (a) The crack-tip process zone for low peeling velocity is very complex involving cavity growth, filament formation and breaking of filament followed by relaxation. (b) At high peeling velocity most of the energy dissipation occurs in a region in front of the crack tip (red hatched region), while the crack-tip process zone is simpler.

and shorter filaments, or even in the absence of filament formation. This will reduce the crack propagation energy. However, as the peeling velocity increases further, the effective interfacial energy to break the rubber-substrate bonds will increase due to viscoelastic deformation of the rubber in front of the peeling edge (red hatched area in

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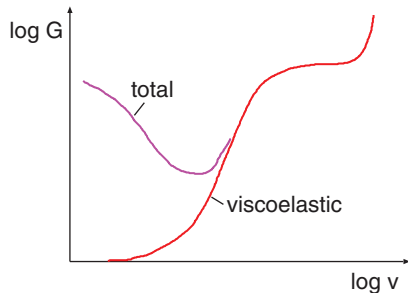


Fig. 2: (Colour on-line) The crack propagation energy G as a function of the peeling velocity v . At low peeling velocity the crack-tip process zone is very complex (see fig. 1(a)), and a lot of energy is dissipated in the crack-tip process zone resulting in a large $G(v)$. At high peeling velocity the crack-tip process zone is simpler but now a lot of energy is dissipated in front of the crack tip as a result of viscoelastic deformation of the rubber (red hatched area in fig. 1(b)).

fig. 1(b)) [8,9]. Thus for large enough peeling velocity the $G(v)$ function will increase as shown in fig. 2. In the region where $G(v)$ is a decreasing function of v , peeling instabilities may occur. This requires some elasticity in the system, *e.g.*, given by the tape itself or by some external spring. In this case, even if the “free” end of the effective spring is pulled at a constant velocity, the adhesive tape may peel in a non-steady way. Non-steady peeling (which we refer to as stick-slip) results in regions of smooth and rough polymer surfaces. We emphasize that the origin of the non-steady motion (or crack-tip instability) is related to the decrease of the crack propagation energy $G(v)$ with increasing crack-tip velocity, which is known to occur in some intermediate velocity range $v_0 < v < v_1$ (where v_0 and v_1 depends on the temperature).

Elastically very soft solids tend to exhibit very small roughness at a short length scale. This is due to the surface tension which will act to reduce the surface area. One extreme case is a fluid, where the surface is perfectly smooth except for thermally excited capillary waves. It is easy to estimate the length scale where the influence of the surface energy on the surface roughness becomes important. Assume that a cubic volume element is cut out of an elastic block with the Young’s modulus E . This volume element will deform in order to reduce the surface energy. If the shape changes from cubic to close to spherical, the surface energy will change by an amount which scales like $\sim R^2\gamma$ while the elastic energy stored up in the solid volume element will scale as $\sigma\varepsilon R^3 = ER^3$, where we have used that the stress $\sigma = E\varepsilon$ and the strain $\varepsilon \approx 1$. The volume where the strain is of order unity scales as R^3 . Thus, if $R^2\gamma > ER^3$ or $R < \gamma/E$, then the surface energy will strongly deform the block.

Consider now instead the surface roughness on a semi-infinite solid. If $h_0(\mathbf{x})$ denotes the initially produced unrelaxed surface profile, and if we write

$$h_0(\mathbf{x}) = \int d^2q h_0(\mathbf{q}) e^{i\mathbf{q}\cdot\mathbf{x}},$$

then the relaxed surface profile (*i.e.*, the surface profile smoothed by the influence of the surface tension γ) will be [10]

$$h(\mathbf{x}) = \int d^2q \frac{h_0(\mathbf{q})}{1 + (2\gamma/E)q} e^{i\mathbf{q}\cdot\mathbf{x}}. \quad (1)$$

The power spectrum $C_0(q)$ (we assume isotropic roughness so that the power spectrum only depends on the magnitude $|\mathbf{q}| = q$ of the wave vector) of the initial surface is given by

$$\langle h_0(\mathbf{q})h_0(\mathbf{q}') \rangle = C_0(q)\delta(\mathbf{q} - \mathbf{q}'), \quad (2)$$

and of the relaxed surface

$$\langle h(\mathbf{q})h(\mathbf{q}') \rangle = C(q)\delta(\mathbf{q} - \mathbf{q}'). \quad (3)$$

Combining (1)–(3) gives

$$C(q) = \frac{C_0(q)}{[1 + (2\gamma/E)q]^2}. \quad (4)$$

Thus, the surface roughness with wave vector $q = 2\pi/\lambda$ will be smoothed by the surface tension if $q > E/\gamma$. For the soft rubber layer used on adhesive tape $E \approx 10^4$ Pa and $\gamma \approx 0.1$ J/m² giving $q > 10^5$ m⁻¹. Thus we expect strongly reduced short-wavelength roughness on pulled adhesive tape. We note that the viscoelastic modulus of rubber-like materials, and in particular the soft rubber used for adhesive tape, depends on time, and becomes smaller the longer the time is. In the present case this implies that the smoothing effect will increase with increasing time after peeling, and the wave vector $q \approx E(t)/\gamma$ which separates the relaxed from the unrelaxed surface roughness, will decrease with increasing time.

We have studied the surface topography of the rubber surface of peeled adhesive tape using an optical method and Atomic Force Microscopy (AFM). The optical surface profiles were acquired using a scanning white-light interferometer (Zygo NewView 6k; Zygo Middlefield, CT, USA) with 5× and 50× objectives at ambient conditions [11]. The AFM measurements were performed in the non-contact mode with a commercial AFM (Veeco Multimode, model MMAFM-2). The AFM was operated in ambient condition in tapping mode using Al-coated cantilevers from Nanosensors (model PPP-NCLR-50). Three different cantilevers were tested with resonance frequencies between 156.50 and 159.50 kHz.

In fig. 3 we show the surface topography $h(\mathbf{x})$ measured in the smooth and rough surface area using the optical method. The root-mean-square (rms) film roughness, including roughness with wavelength $\lambda < 1$ mm is 0.05 μm on the most smooth surface area and 0.5 μm on the most rough surface area, but the fluctuations between the highest and lowest points on the surfaces are several times higher than the rms-roughness values. Nevertheless, the thickness of the rubber film (of order 20 μm) is much larger than the roughness amplitude.

In fig. 4 we show the surface roughness power spectrum calculated from the measured surface topography, $h(\mathbf{x})$,

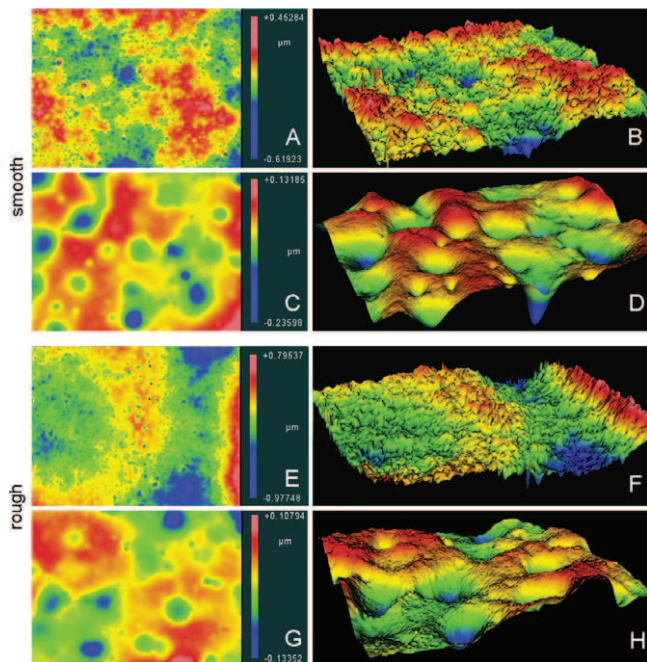


Fig. 3: (Colour on-line) Surface profiles of the adhesive side of the scotch tape obtained by the use of white-light interferometer. A–D: smooth part (fast peeling). E–H: rough part (slow peeling). A,B,E,F: low magnification ($\times 5$) (surface area $1 \text{ mm} \times 1 \text{ mm}$). C,D,G,H: high magnification ($\times 50$) ($0.1 \text{ mm} \times 0.1 \text{ mm}$).

for several differently peeled surfaces. It appears as if $C(q)$ exhibit a power law in q (which gives a straight line on a log-log scale) in all the studied (smooth and rough) surface areas, *with the same slope* in all cases but with different root-mean-square roughness. For a self-affine fractal surface $C(q) \sim q^{-2(1+H)}$, where the fractal dimension is related to the Hurst exponent H via $D_f = 3 - H$. The dashed line in fig. 4 has a slope corresponding to $D_f = 2.3$, which is typical for surfaces prepared by fracture but unexpected in the present case since if the fracture process initially generated a profile with a power spectra $C_0(q)$ one would expect the power spectra of the relaxed profile (for $q > E/\gamma$) to be $C(q) \sim C_0(q)/q^2$. In particular, one would expect a change in the slope of the $\log q - \log C$ relation for $\log q \approx \log(E/\gamma) \approx 5$, which is not observed (see fig. 4).

The results presented above may be very important not only for gaining insight into the processes involved in the interfacial bond-breaking but also in other phenomena observed during peeling adhesive tape. One of us has recently suggested that the surface roughness on adhesive tape may be involved in an important way in the the emission of high-energy photons which has been observed during peeling adhesive tape [12]. In general, it is well known that the relative motion between two contacting solids can produce light, called triboluminescence [13]. For example, opening an envelope in a dark room usually result in flashes of blue light from the (pressure-sensitive

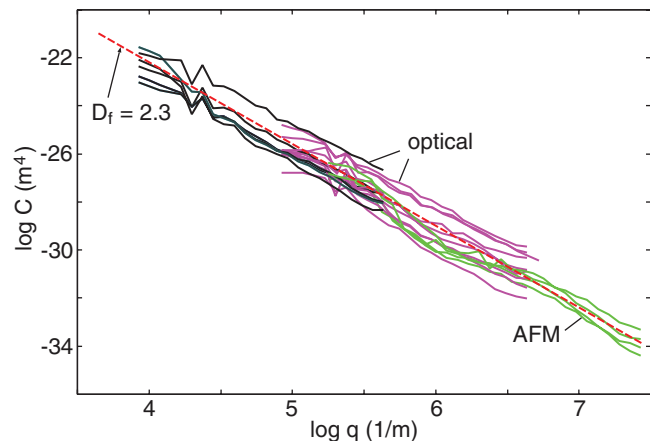


Fig. 4: (Colour on-line) The logarithm of the surface roughness power spectrum C as a function of the logarithm of the wave vector for a peeled adhesive tape surface. There are different regions on the tape surface with low and higher surface roughness. The topography was measured using an optical method (at low magnification ($\times 5$) over $1 \text{ mm} \times 1 \text{ mm}$, black lines, and at high magnification ($\times 50$) over $0.1 \text{ mm} \times 0.1 \text{ mm}$, pink lines) and using AFM ($50 \mu\text{m} \times 50 \mu\text{m}$, green lines). The root-mean-square surface roughness, including roughness with wavelength $\lambda < 1 \text{ mm}$, is $0.05 \mu\text{m}$ on the most smooth surface area and $0.5 \mu\text{m}$ on the most rough surface area.

rubber) adhesive interface. Recent experiments [14] have shown that photons with energies up to $\sim 100 \text{ keV}$ are produced during the peeling of adhesive tape. The X-ray pulses were correlated with stick-slip peeling events.

This generation of high-energy photons is believed to be related to charge separation at the opening crack tip. In ref. [12] one of us has proposed that the emission of high-energy photons involves surface roughness and surface diffusion of electrons or ions, resulting in the concentration of charge at the tips of high asperities, and to electric-field enhancement, which facilitates the discharging process which results in the high-energy photons. In ref. [12] it was shown that the enhancement of the electric field is necessary not just for triggering the avalanches but also for explaining the observed maximum in the emitted photon energy observed in ref. [14]. The information about the surface roughness gained in this paper may be used when calculating electric-field enhancements relevant for the interpretation of the results of ref. [14].

Additionally, information about roughness generated by peeling might be important for understanding the adhesion behavior in multiple successive attachment-detachment cycles, when the tape is repeatedly used. In order to stabilize adhesion in successive adhesive cycles, a specific roughness might be built in during the production process.

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