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Human ability to discriminate surface chemistry by touch†

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The sense of touch is mediated by the interaction of a soft material (*i.e.*, skin) with the texture and chemistry of an object's surface. Previous work designed to probe the limits of tactile perception has been limited to materials with surface asperities larger than the molecular scale; such materials may also have different bulk properties. We demonstrate in a series of psychophysical experiments that humans can discriminate surfaces that differ by only a single layer of molecules, and can "read" patterns of hydrophobicity in the form of characters in the ASCII alphabet. We design an apparatus that mimics free exploration of surfaces by humans and corroborate the experimental results with a theoretical model of friction that predicts the velocities and pressures that permit discrimination. These results demonstrate that forces produced, while sliding a finger along surfaces, interact with the mechanoreceptors of the skin to allow the brain to discriminate surfaces that differ only by surface chemistry. While we used intentionally simple surface modifications in this study (silanized *vs.* oxidized silicon), these experiments establish a precedent for using the techniques of materials chemistry in psychology. They also open the door for the use of more sophisticated, molecularly engineered, materials in the future.

Conceptual insights

Can humans discriminate between two surfaces that differ by a single layer of molecules at the surface solely with the sense of touch? This paper seeks to answer this question by combining the tools of surface science, psychophysics, and tribology. As material scientists, we are hesitant to touch samples out of fear of damaging or contaminating them. This behaviour, rational as it is, has prevented us from asking fundamental questions pertaining to our sense of touch that can only be answered using the tools of materials science. To date, psychophysical studies have traditionally been designed using "off-the-shelf" materials that differ in multiple properties, which introduce many confounding variables and effects. This paper introduces the methodology of materials science to the toolkit of psychology in order to explore the interface between the human sense of touch and the material world. We found that indeed humans are capable of detecting differences between smooth surfaces that differ only by their topmost layer of molecules (*i.e.*, they have different surface energies). These surfaces are discriminable due to differences in vibrational frequencies generated while sliding. These psychophysical insights are supported using a silicone mock-up of a finger along with a mathematical model.

Tactile perception of an object is influenced by several parameters: its bulk properties (*e.g.*, hardness[1] and thermal conductivity[2]), its surface properties (*e.g.*, roughness[3]), and variables of extrinsic origin (*e.g.*, thin wetting films[4]). When an object is interrogated with a fingertip at a given force and velocity, these properties trigger sensations in the skin[5] as well as the joints of the hand and arm, and as vibrations detected by the ear, to produce tactile images in consciousness. It is known that the skin is capable of registering minute differences in periodic roughness[3] and thermal properties,[2] but the mechanism by which human subjects distinguish objects based only on surface chemistry is not known. Such knowledge is critical in the development of haptic technology using soft, active materials, and would accelerate development of electronic skin,[6] instrumented prostheses,[7] devices for physical therapy, and enhanced robotic surgery.[8] It may also lay the groundwork for tactile artwork[9] and a neurological understanding of tactile illusions.[10]

It is difficult to overstate the importance of the tactile sense in medicine, psychology, and technology. Tactile and variable-friction displays,[11] anti-fouling surface coatings,[12] and advanced haptic interfaces for virtual and augmented reality[13] require knowledge of how the properties of materials are perceived by touch. Additional factors such as the morphology and hydration of the skin also contribute to the "feel" of an object.[4] Elucidating the mechanisms that influence tactile perception of objects in the environment requires control over the properties of materials on the molecular scale. Recent work to establish a connection

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between materials science and the tactile sense focused on, for example, the human ability to discriminate surfaces exhibiting nanoscale differences in micron-scale wavy topographies (*e.g.*, akin to judging which sandpaper is finer, but on the nanoscale), while holding surface chemistry constant.[3] Previous studies to determine the contact mechanics of interfaces between human skin and materials exhibiting different surface chemistries (*e.g.*, glass *vs.* acrylic resin) were confounded by differences in bulk properties (*e.g.*, thermal conductivities and mechanical properties[14]). The effects of surface chemistry alone—with all bulk properties held constant—have not been explored.

Modification of silicon and silicon oxide surfaces using fluorinated alkylsilanes[15] is a ubiquitous approach to control fouling and adhesion in the design of touch screens[12] and microelectromechanical systems (MEMS).[16] Silane monolayers do not affect the bulk properties of the substrate, and thus can be used to isolate the effects of surface chemistry. Systematic control over surface chemistry, and the way it modulates surface forces, may establish a molecular basis—and unveil new limits—of tactile perception. The goal of this work was to test whether humans can discriminate objects based on surface chemistry and to establish a framework for discrimination by touch. That is, to develop a theory—supported by psychophysical and mechanical measurements as well as analytical models—to describe how single layers of molecules give rise to adhesion and friction forces that produce differentiable signals in the brain when sliding or tapping a finger across the surface. If it can be shown that humans possess such sensitivity to surface chemistry, it should be possible to encode information spatially that cannot be detected by any sense other than touch. Moreover, such knowledge might stimulate the development of dynamic, reconfigurable materials that can produce a range of sensations for physical therapy, education, and virtual and augmented reality.

We began by determining whether human subjects could discriminate between near atomically smooth silicon wafers (R_a^{Si} = 0.113 nm) with two different surface chemistries. (I) Hydrophobic: passivated with a fluorinated alkyl silane (''FOTS,'' R_a^{FOTS} = 0.206 nm). (II) Hydrophilic: activated by plasma oxidation (''SiOH,'' R_a^{SiOH} = 0.203 nm). In each of eight trials, subjects (n = 15) were asked to freely explore a set of three surfaces and identify the one dissimilar surface (the ''odd-man-out''[14]) using only their sense of touch (Fig. 1c, top). Between subjects, FOTS surfaces were wiped thoroughly with isopropanol, while SiOH surfaces were wiped with isopropanol and re-treated with oxygen plasma <1 h before human subject experiments. Washing did not affect the contact angle of the FOTS surfaces, while the SiOH surfaces retained a water contact angle of zero for several hours after plasma treatment. While it may seem ''obvious'' that humans could detect the differences between these surfaces based on our intuitive sense that ''stickiness'' increases with surface energy, in reality discriminating these surfaces is not easy, and, in pilot experiments, some subjects could not discriminate between them at all.

We used generalized mixed-effects modeling (GMM; see Methods) to quantify subjects' accuracy of discrimination. Subjects correctly identified the dissimilar surfaces significantly more often than predicted by chance (Fig. 1d, top bar, mean accuracy = 71.7%; Wald Z test, $P < 0.0001$). However, we found a trending inverse correlation between accuracy and moisture of the skin (Fig. 1e; Wald Z test, $P = 0.067$). It should be noted that skin moisture levels increase drastically when contacting impermeable surfaces on the order of 10 s due to the occlusion of eccrine sweat from the glands of glabrous skin, *i.e.* the hairless skin found on palm and fingers of the hand and the bottoms of the feet.[17] Therefore, moisture measurements taken before engagement with the surface may not always serve as a robust predictor of accuracy during extended free exploration. To eliminate the possible confounding effect of hydration and capillary forces (*i.e.*, to isolate the effect of van der Waals forces), the experiment was repeated with the wafers submerged in deionized water. In this ''wet'' condition, subjects (n = 15, same subjects as the ''dry'' experiment) could still identify the dissimilar surface significantly more often than predicted by chance (Fig. 1d, middle bar, mean accuracy = 84.17%; Wald Z test, $P < 0.0001$). In fact, subjects were significantly more accurate in the ''wet'' condition than in the ''dry'' condition (Wald Z test, $P < 0.05$). However, we cannot eliminate a possible training effect: all subjects in the ''wet'' experiment had previously experienced the discrimination task in the ''dry'' experiment, so the increase in accuracy might have resulted from practice. It is clear, nevertheless, that conditions unique to the ''dry'' experiment were not necessary to perform the discrimination task. This experiment suggests that differences in capillary adhesion between the two surfaces are not necessary to discriminate between surfaces.

Verbal descriptions of the surfaces by the subjects as being ''smoother'', ''stickier'', and ''slipperier'' strongly suggested that friction played a role in the ability of the subject to discriminate between surfaces. It is possible, however, that adhesive forces, felt at the first moment of touching the surface or lifting the finger off the surface, also played a role. To isolate possible effects of adhesion of the finger to the surface (*i.e.*, tackiness) from those of friction, the experiment was repeated, but subjects were instructed to tap the surfaces rather than explore them freely. Subjects (n = 14, 8 new subjects) could still identify the dissimilar surface significantly more often than predicted by chance (Fig. 1d, bottom bar, mean accuracy = 56.25%; Wald Z test, $P < 0.01$), but significantly less often than in the free exploration conditions (*vs.* Wet: Wald Z test, $P < 0.0001$; *vs.* Dry: Wald Z test, $P < 0.01$). It is thus clear that the subjects could perceive molecular differences in surfaces based on adhesion alone, but were significantly more accurate when given the chance to explore surfaces freely (by sliding) rather than restricted to tapping alone. Higher accuracy in free exploration over tapping alone suggests that friction during sliding acted as the primary cue for successful discrimination.

The chemical nature of the interface between the skin and the surface is highly complex and varies between individuals and over time. A finger—even after washing—will deposit eccrine secretions and exfoliated skin. The deposited material consists mostly of inorganic ions, amino acids, and lipids.[18] Free exploration of

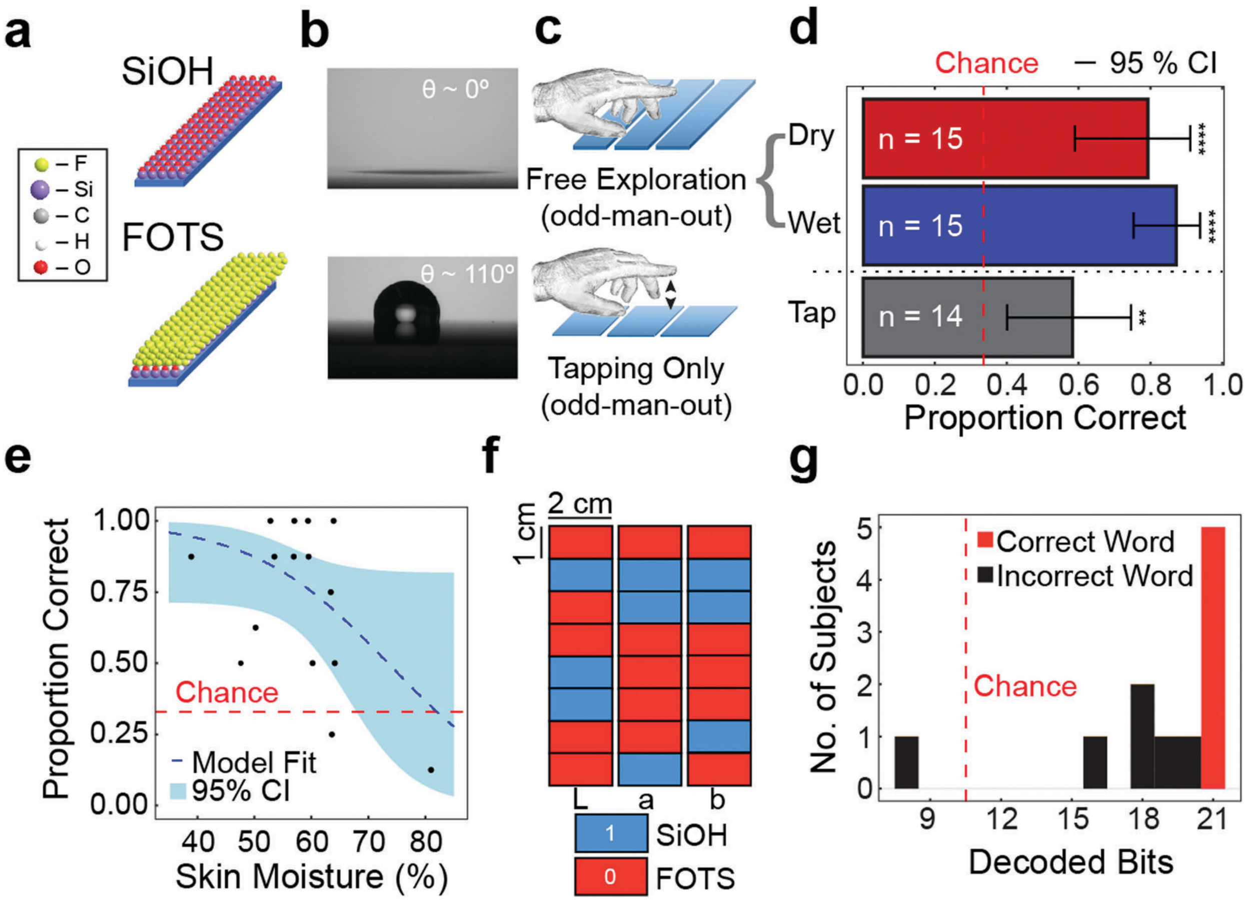

Fig. 1 Summary of psychophysical results. (a) Schematic diagram of SiOH (top) and FOTS (bottom) surfaces. (b) Contact angles of 2 μL water droplets on SiOH (top, static water contact angle = 0°) and FOTS (bottom, static water contact angle = 110°) surfaces. (c) Free exploration (top) and tapping only (bottom) in an "odd-man-out" test. (d) Behavioural results of discrimination experiments. Data are mean accuracy and 95% confidence interval of the GMM intercept term (see Methods). $^{**}P < 0.01$, $^{****}P < 0.0001$. (e) Subject accuracy (y-axis) in the "dry" condition as a function of finger pad moisture level (x-axis). Red dashed line depicts chance performance. Data are individual subject performance (points), GMM fixed effect (blue dashed line), and 95% confidence interval on fixed effect (see Methods). (f) Schematic diagram of "molecular braille" corresponding to rectangular regions of silicon wafers (2 cm × 8 cm) using 1 cm SiOH and FOTS patterned segments to spell the word "Lab" over three separate wafers. (g) Plot showing the distribution of successfully decoded bits among subjects (21 successfully decoded bits corresponds to the correct word). Red dashed line depicts chance performance.

an initially clean surface means that subjects could pass over a section of the surface that was previously traversed. We thus acknowledge the possibility that secretions could have influenced the ability of human subjects to discriminate the surfaces. We note, however, that these secretions are always present between the skin and the surface whether or not they were on the surface in a previously traversed region. Our measurements and observations suggest that while pre-deposited material may have played a role in the ability of human subjects to discriminate surfaces, the chemistry of the native surface is sufficient. To support this claim, a single subject was asked to perform the odd-man-out test, but was restricted to swiping only previously unexplored regions of each sample. During this experiment—restricted exploration, as opposed to free exploration—the subject correctly identified the odd-man-out in five of eight trials. Following this experiment, atomic force microscopy and optical microscopy were performed to visualize the deposition on each surface after a single swipe with a length of 2.5 cm (Fig. S3, ESI†). FOTS surfaces exhibited consistent deposition from the beginning to end of a single swipe, while SiOH surfaces showed less deposition (or simply less smearing of material deposited initially). Contact angles measured on touched regions of each surface maintained a contact angle of zero for SiOH and only a minor increase in the advancing contact angle ($\theta_A = 115°$ initially, $\theta_A = 118°$ after touching) and a decrease in the receding contact angle ($\theta_R = 92°$ initially, $\theta_R = 79°$ after touching) of FOTS surfaces. Unfortunately, labile material on the surface of the skin is unavoidable and depends on the hydration, surface temperature, and level of keratinization of the skin of each subject. This level of variability makes the degree of human sensitivity to surface chemistry as revealed by the psychophysical experiments even more remarkable.

To test the ability of subjects to distinguish regions of hydrophobicity and hydrophilicity with lateral resolution, we asked subjects to "read" sequences of hydrophilic ("SiOH") and hydrophobic ("FOTS") patches (1 cm long) on a surface representing "1" and "0" bits of the ASCII alphabet, a form of tactile communication akin to braille. Fig. 1g shows that 10 of 11 subjects decoded bits of the word "Lab" with accuracy significantly better than chance (binomial tests, all $P < 0.05$) and identified each letter in 4.5 min on average. We note that subjects were aware that the three strings of eight bits combined to form a word rather than a random sequence of letters, which allowed subjects to self-correct for errors during the experiment. We did not try to test the limit of lateral resolution, but we expect that the accuracy would degrade if the lateral size of the hydrophobic and hydrophilic patches were significantly smaller than 1 cm. We also note that while the FOTS monolayer in principle has a step height of ~1 nm, it is the differences in surface energy (mediated by chemistry), rather than the height, that were being detected by the subjects. (Though remarkably, human subjects can perceive periodic relief features that differ in amplitudes as small as 10 nm.)[3]

The next task was to link subjects' abilities to discriminate between surfaces to physical phenomena. Audible sounds produced at various points during free exploration of the surfaces by the subjects were consistent with stick-slip friction. We recorded the sounds (Fig. 2a) and converted them to the frequency domain using a Fourier transform (Fig. 2b). The two surfaces were observed to differ in the sounds produced when interrogated at approximately the same velocity and normal force, as the FOTS surface produced two peaks at 101 and 389 Hz, while the SiOH surface produced one prominent peak at 236 Hz. Vibrational frequencies in this range are detected by the Pacinian corpuscles in the deep dermis, while stretching and movement of the skin (*e.g.*, by sliding the finger along the surface) are registered by the Ruffini endings and Meissner corpuscles.[5] While samples can produce different sounds, most subjects used a light touch that did not produce sounds loud enough to be detected (subjects also wore noise-cancelling headphones that limited auditory cues).

It is commonly accepted to quantify surfaces based on the static and kinetic coefficients of friction, even though these coefficients are highly dependent on the testing conditions,[19] and ignore dynamic instabilities like stick-slip phenomena. The fact that subjects were more accurate in free exploration *versus* tapping alone would make it tempting to attribute the ability of the subjects to discriminate between the two surfaces to a difference in friction coefficients, considering the static friction coefficients for FOTS and SiOH are quite different (0.13[20] *versus* 0.44[16]). However, we set out to take a closer look using a mechanical model system since the actual friction forces could be identical under many conditions (*i.e.*, some combinations of normal force and velocity may actually produce similar friction forces).

To investigate the effects of a subject's sliding velocity and applied force on discriminability, we built a custom apparatus drawn schematically in Fig. 3a. This apparatus comprised a force sensor attached to a ''finger'' made from a block of poly(dimethylsiloxane) (PDMS) with an oxidized surface to reduce its viscoelastic tack. We mimicked free exploration by testing a range of swiping velocities and normal forces and calculated the cross-correlation between the force traces for the two surfaces. A strong correlation suggests that the surfaces would not be discriminable, while a weak correlation would suggest that the surfaces would be. Since human subjects interrogate objects using free exploration (and unconsciously vary the velocity and force), it is possible that surfaces are only discriminable given certain combinations of velocity and force. Subjects could therefore pass through regions of a hypothetical parameter space of discriminability and non-discriminability multiple times in a single engagement with a surface.

A complete force *vs.* time trace obtained by the model finger (PDMS block) sliding on a surface is shown in Fig. 3b. The traces of force *vs.* time had oscillations characteristic of stick-slip behaviour. The first peak is always ignored and the force traces used in the analysis are in the boxed region, labelled I, II and III. Fig. 3c and d highlight experiments from 2 of the 16 combinations of velocity and force chosen on the basis of whether or not the surfaces were discriminable by cross-correlation. The left-hand column (Fig. 3c, e, and g) represents a discriminable case, while the right-hand column (Fig. 3d, f, and h) represents a non-discriminable case. In Fig. 3c (v = 2.5 mm s^{-1} and M = 0 g), the force traces of PDMS fingers pulled on FOTS and SiOH-treated surfaces are visually different. M refers to the mass added to the finger, which has a deadweight of 5 g. To avoid possible interference from deposition of unpolymerized material from the PDMS

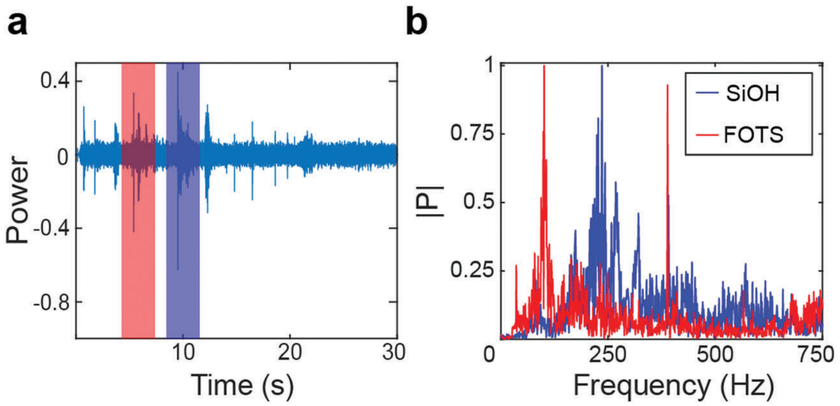

Fig. 2 Audible evidence for stick-slip friction. (a) Analysis of raw audio signal of finger sliding across FOTS (red box) and SiOH (blue box) surfaces. (b) Plot of FFT power analysis of raw audio signals for FOTS (red line) and SiOH (blue line) surfaces.

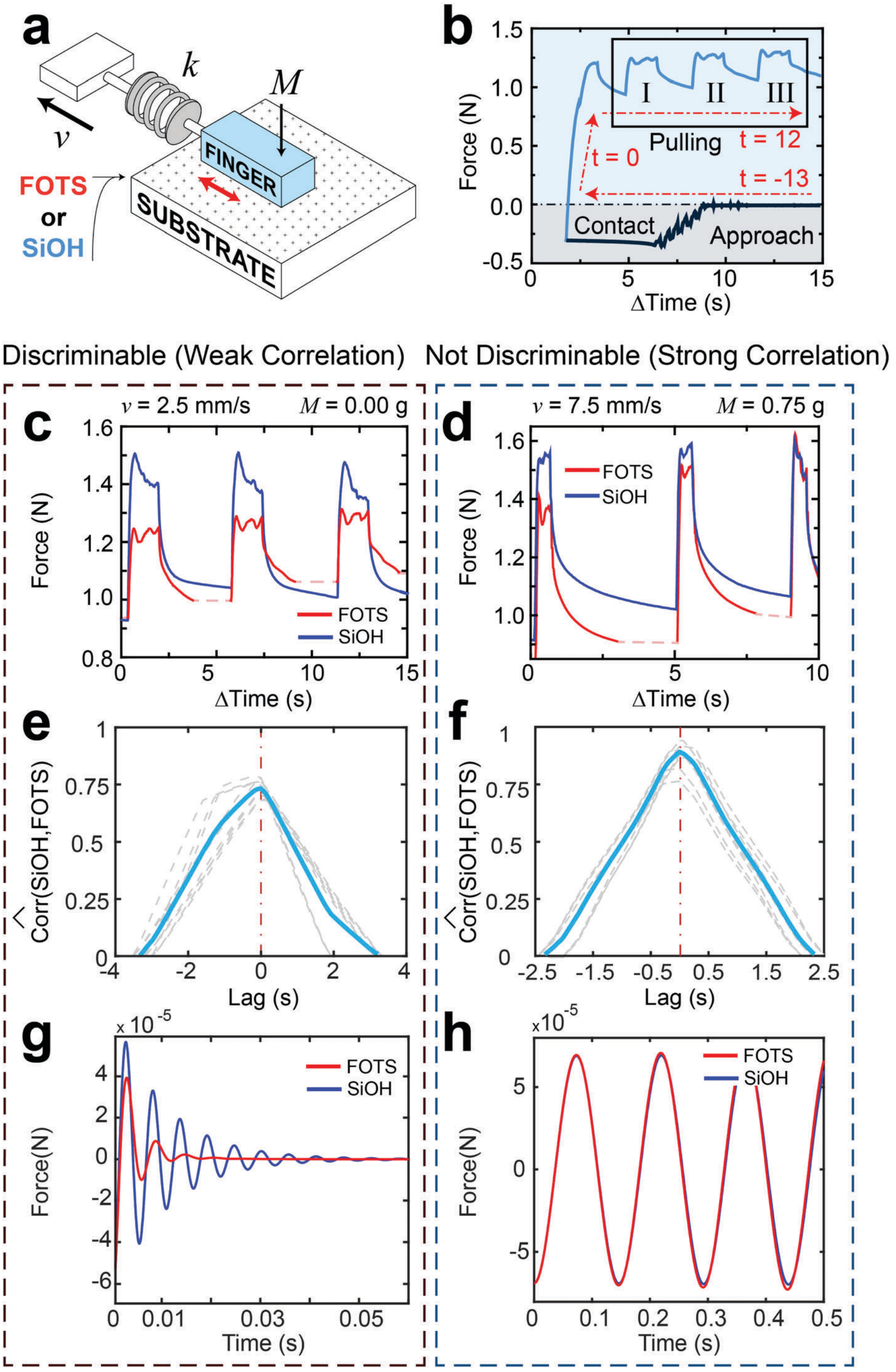

Fig. 3 Friction measurements of PDMS on silicon wafers with FOTS or SiOH surfaces. (a) Schematic diagram of the apparatus to measure the friction force of a model finger (PDMS block). (b) Typical profile of the loading and pulling phases. The first pull after the approach phase was ignored, and then the subsequent three pulls (I–III) were measured. (c and d) Representative force *vs.* time traces of the PDMS block on FOTS and SiOH for $v = 2.5$ mm s^{-1}, applied mass = 0 g in (c) and $v = 7.5$ mm s^{-1}, applied mass = 0.75 g. Force traces of samples tested on FOTS have been shifted along the x-axis for easier visual comparison to SiOH. (e and f) The normalized correlation coefficient of the force *vs.* time traces in (c and d) shown in (e and f), respectively. The solid line represents the average correlation, and the grey, dashed lines represent the individual correlations. The dashed-red line is a visual guide for symmetry about the x-axis. (g and h) The oscillations in force due to sliding friction for a block on surfaces treated with FOTS and SiOH as predicted by the friction model.[22]

block, we used a new region of the SiOH or FOTS substrate for each measurement. There is a prominent initial spike (stiction) in force[21] on the SiOH surfaces, while the FOTS surface appears to oscillate evenly in force. In Fig. 3d ($v = 7.5$ mm s^{-1} and $M = 0.75$ g), the traces are visually indistinguishable. We calculated a normalized cross-correlation to quantify the similarity in force traces. In Fig. 3e, the cross-correlation is asymmetric about lag = 0 with a peak correlation value around 0.75 while the cross-correlation in

Fig. 3f is more symmetric and the peak correlation is higher at approximately 0.9.

We modelled the friction forces using the simplest model that accounts for stick-slip phenomena.[22] This model introduces the concept of a "state" variable (θ), which accounts for how the friction force varies with the local velocity and displacement of the block, and the time-dependent friction coefficient.[23] Treating the finger as a rigid block connected through a spring to a driver and sliding along one axis, the friction coefficient and the state variable are given in eqn (1) and (2):

$$\frac{F_{\parallel}}{F_{\mathrm{N}}} = \mu = \left(\mu_{\mathrm{o}} + \theta + A\ln\left(\frac{v}{v_{\mathrm{o}}}\right)\right) \tag{1}$$

$$\frac{\mathrm{d}\theta}{\mathrm{d}t} = \left(-\frac{v}{D_{\mathrm{c}}}\right)\left(\theta + B\ln\left(\frac{v}{v_{\mathrm{o}}}\right)\right) \tag{2}$$

where $F_{\parallel}$ and F_{N} are the parallel force and normal force on the block, t is time, μ is the friction coefficient, v is the velocity of the block, v_{o} is the motor drive velocity, and A, B, μ_{o} and D_{c} are the friction parameters unique to each material (extracted by plotting μ *versus* v). Oscillations that arise from stick-slip phenomena are shown in Fig. 3g and h. In Fig. 3g, we see that the oscillations between the substrates are distinct, while in Fig. 3h, the oscillations overlap both in magnitude and frequency.

To compare the simple friction model to the experimental output, we created two scoring matrices (Fig. 4). A value of "1" (green) signifies that the substrates exhibit differences in friction forces (and presumably, human perception) while a value of "0" (red) signifies similarity. For the experimental results, we picked a weighted combination of the normalized area under the curve and the normalized skew of the correlation plot, while, for the mathematical model, we picked a weighted combination of the differences in the number of zero crosses and the differences in magnitude. These weighted combinations give rise to a combined score for the experiments (Fig. 4a) and a predicted one from the model (Fig. 4b), which serve as discrimination matrices. The general trend in both appears to be a sweeping, top-left to bottom-right range in high discriminability, with the largest differences between the substrates at low masses and low velocities. These findings confirm the need to model both experimentally and mathematically the connection between sliding friction and tactile perception,

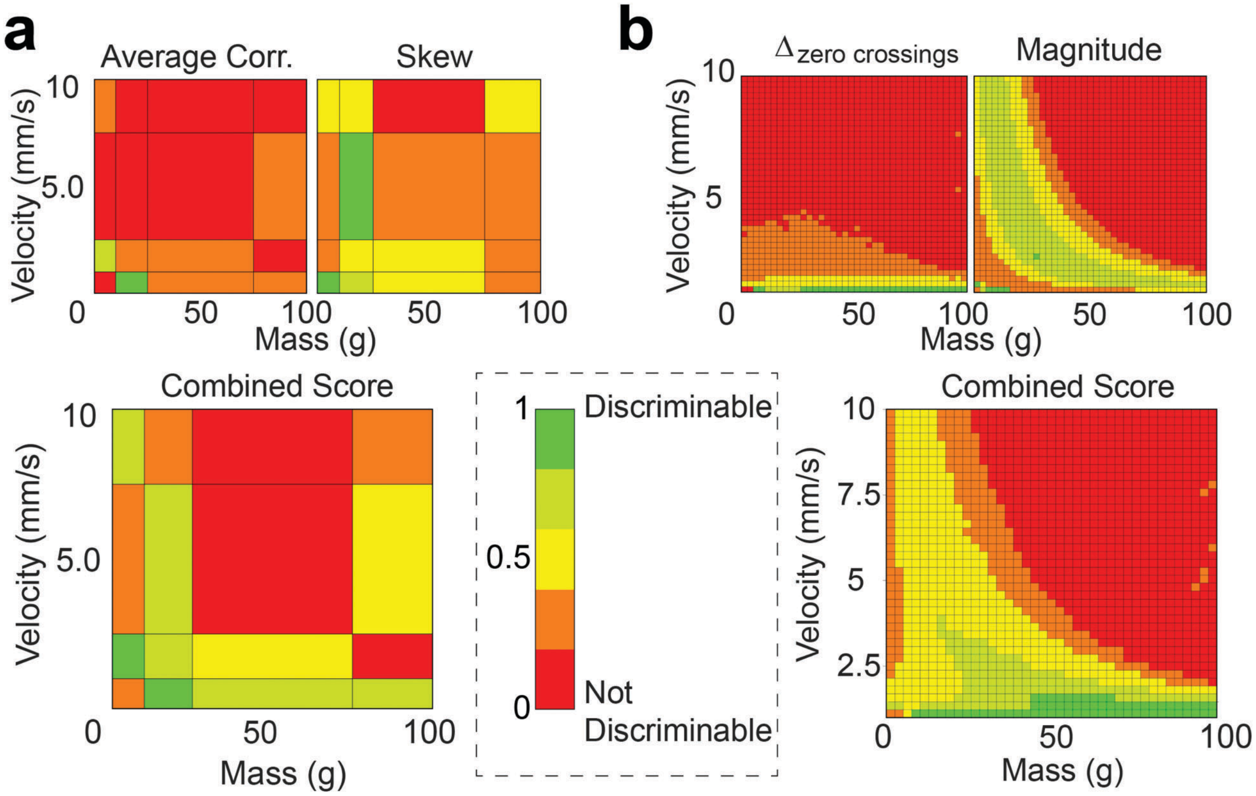

Fig. 4 Visualized discriminability score of FOTS and SiOH surfaces from experiments and theory. As shown in the legend with the dashed border, a value of 1 (green) means the FOTS and SiOH surfaces are discriminable, whereas a value of 0 represents surfaces that are not discriminable. (a) Experimental results of the cross-correlation when sliding a PDMS block on FOTS and SiOH and two metrics used to evaluate the cross-correlation which were the average value of the cross-correlation, normalized by a maximum cross-correlation value and the skew of the cross-correlation, normalized by the largest skew value in the dataset. The combined score shows the velocities and masses where force traces of FOTS and SiOH are discriminable or not. (b) Theoretical oscillations in force due to sliding friction for surfaces on FOTS and SiOH. The first metric here is "$\Delta_{\text{zero crossings}}$", which compares the difference in frequency of oscillations on FOTS and SiOH by quantifying the changes in direction (from positive to negative). The second metric is "Magnitude", which is the percentage of the time where the amplitude of oscillations (force) in the friction traces of FOTS and SiOH varies by at least a factor of five.

which is not predictable simply from knowledge of the friction coefficient.

Conclusions

Our results reveal a remarkable human ability to discriminate surfaces based only on surface chemistry: untrained individuals can quickly home in on the normal forces and sliding velocities required to distinguish surfaces that differ by a single layer of molecules. Subjects can use this ability to decode information—*i.e.*, digital bits and possibly also shapes—that is undetectable by every sense except touch. While adhesion does allow subjects to discriminate between FOTS and SiOH surfaces (as revealed by tapping experiments), the primary mechanism that permits this ability appears to be unequal vibrational frequencies arising from stick-slip friction behaviour triggered by different forces and velocities of interrogation. Interestingly, knowledge of the coefficient of static friction appears to be an insufficient criterion for discriminability. That is, objects with different surface chemistries can "feel" the same with many combinations of forces and velocities, according to the results of both a purpose-built apparatus and an analytical model. Taken together, these results elucidate the limits of the tactile sense and highlight the need for more interdisciplinary research, in which tactile perception (including its neural and physiological aspects) is investigated using the tools of modern materials science.[24] Better understanding of the relationship between physical properties and human touch perception could spur the development of new, stimulus-responsive materials[25] for haptic feedback and enhanced human-machine interfaces.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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