Research Highlight #1500

Tevis Jacobs, Ph.D.

Whiteford Fellow and Associate Professor, University of Pittsburgh

Professor Tevis Jacobs describes his research as all fundamentally addressing the same question: how do we understand interactions at surfaces? His group at the University of Pittsburgh approaches this question at all scales from the nanoscale to the macroscale.

Origin Story: Nanoscale Wear

Exciting discoveries about nanoscale wear

Typically, wear testing is conducted on a macro scale. For his dissertation work, Jacobs scaled wear testing down to the nanoscale. With implications especially for nanotechnology applications, Jacobs showed that nanoscale wear of silicon occurs in an atom-by-atom fashion that could be described and predicted by reaction rate theory. He published this work in the journal *Nature Nanotechnology* in 2013[1].

An interest in roughness is launched

After spending hundreds of hours analyzing nanoscale surface interactions inside of a transmission electron microscope (TEM) during his Ph.D. and postdoc work, Jacobs knew that he wanted to understand the implications of surface effects for real-world devices. With his group at the University of Pittsburgh, he attempts to study this in two ways: exploring the atomic-scale physics of nanoscale contacts[2]; and investigating how surface topography links that to macroscale devices[3]. Though he enthusiastically admitted that he could discuss either topic at length, he chose to focus on the nanoscale studies in this highlight.

Projects as Principal Investigator: Nanoscale Contact Area and Conductance

Do conventional contact area models work?

Solid mechanics models can tell us what contact area to expect when a sphere of any size rests on a surface. But many continuum mechanics models break down at the nanoscale, so Jacobs wanted to test conventional assumptions. By applying a load in-situ to samples within the TEM, he and his group didn't need to assume – they could see and measure. They did not see what the models predicted.

Conventional models underpredicted the actual contact area at the nanoscale by, in some cases, up to 100%. This is very important because usually researchers cannot see their nanoscale tip (e.g., in AFM applications or nanodevices) and must use established models to assume a contact area in subsequent equations. One question remained for the Jacobs Group though: why were the established models underpredicting?

ABOUT THE RESEARCHER

Dr. Tevis Jacobs is a William Kepler Whiteford Faculty Fellow and Associate Professor in the Department of Mechanical Engineering and Materials Science at the University of Pittsburgh.

- PhD, Materials Science and Engineering, University of Pennsylvania, 2008 - 2013
- MSc, Materials Science and Engineering, Stanford University, 2004 - 2006
- MPhil, Computer Modeling of Materials, Churchill College, Cambridge University, 2003 - 2004
- BSc, Mechanical Engineering, also Materials Science and Engineering, University of Pennsylvania, 1999 – 2003

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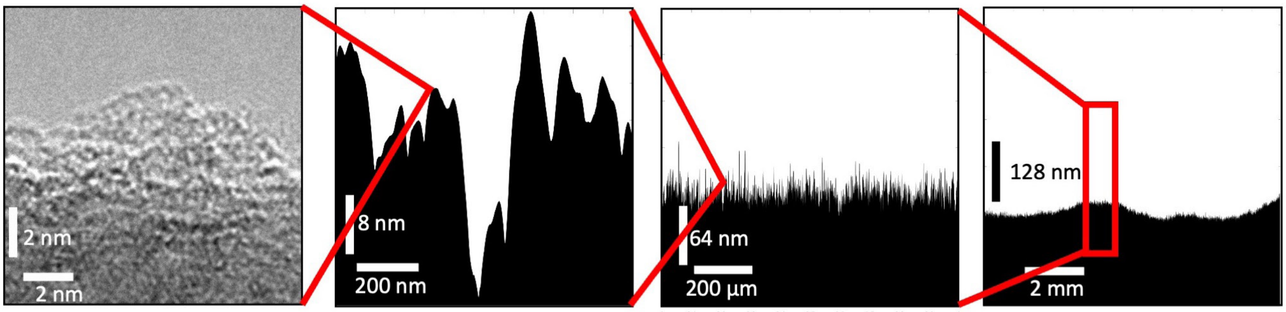
> "[My] early work launched my interest in the effect of roughness on properties. Surface topography allows us to link together different size scales."



After confirming that their findings weren't due to irreversible plastic deformation blunting the tip, Jacobs and his Ph.D. student Sai Bharadwaj Vishnubholta were determined to figure out the mechanisms. Eventually they showed how reversible plasticity occurring within the probe tip served to increase the contact area. Dislocations formed and propagated, but when the load was removed, they annihilated at the surface. Functionally, this meant that the contact was much larger than predicted by elastic models, but with reversibility that was not predicted by plastic models. They published their findings in *Nanotechnology* in 2019[4].

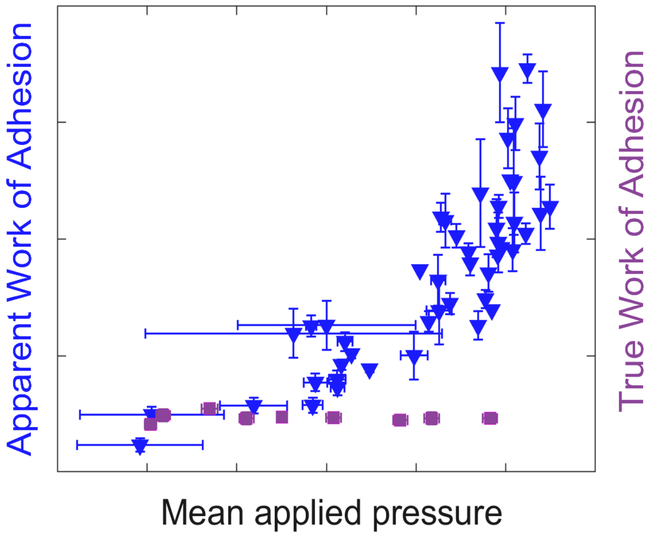
Revising the nanoscale conductance model

Knowing the true contact area at the nanoscale, Jacobs began to extend that insight to conductance at the nanoscale. Just like there are models for calculating contact area, there are also models for predicting conductance using the contact area. Just like the contact area models, the in-situ TEM testing allows the determination of whether these conductance models agree with direct nanoscale observation?

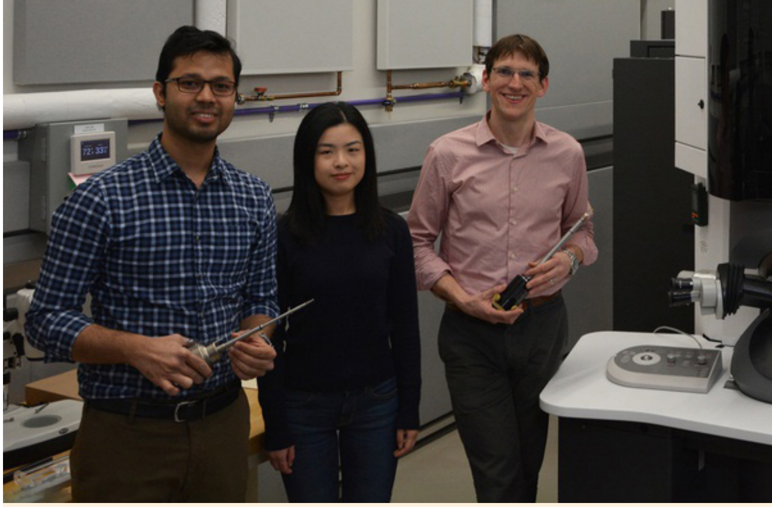
If conductance models are not accurate, it would affect calculations converting measured current to contact area, for example in applications such as AFM and nanoscale switches. Once again using in-situ testing, Jacobs and Vishnubholta examined the true conductance across the tip during loading while also measuring the contact area. Even knowing the true contact area, the typically-applied ballistic or diffusive conduction models did not accurately describe conductance across these platinum contacts.

The idealized view of the interface failed to account for monolayers of insulating surface species, even on contacts of noble metals like platinum. Due to the presence and persistence of these layers, it was more accurate to apply tunneling theory for electron transport, rather than ballistic or diffusive conduction. They published this work in *Nanotechnology* in 2019[5], along with a supporting simulation-focused 2019 manuscript in *Nanoscale*[6].

"A lot of our work has been asking that question: do these conventional models work? If they do work, great, why do they work? Why do continuum models apply all the way to the atomic scale? And if they don't work, it's not enough to just say that they break down, you need to figure out how they break down? Why? And what models should replace them?"



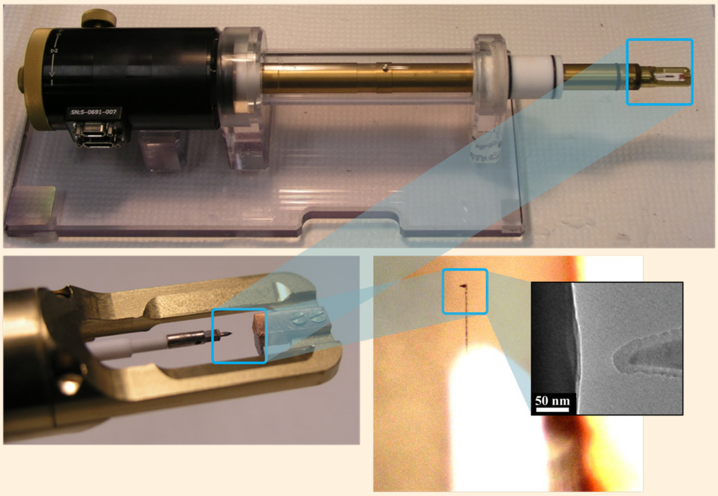
"This ended up being a story about how classical models can lead us astray at the nanoscale, and more importantly why."



Understanding “load-dependent” work of adhesion

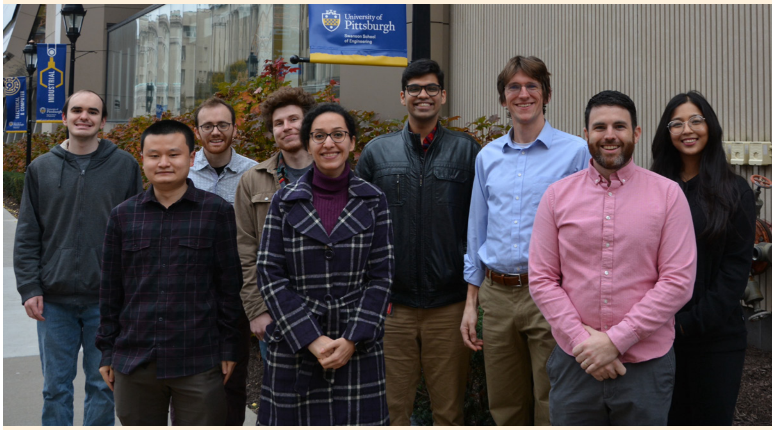
Jacobs and his Ph.D. student Andrew Baker moved on to the next property to investigate with direct observations of nanoscale contacts: the adhesion energy between two surfaces. In general, an incredibly common method for measuring adhesion energy (also called work of adhesion) relies on AFM-based adhesion tests. By measuring the force required to separate a tip of known radius from a surface, and by relying on continuum models, there is a simple expression for computing adhesion energy. But how accurately do these continuum models apply for these nanoscale contacts?

Conventional models assume that there is a constant adhesion energy pulling surfaces together. By contrast, recent work on nanoscale contacts[7, 8, 9] has shown that adhesion force, and the adhesion energy computed from this force, is not constant, but rather varies with load. In 2022, Jacobs and Baker investigated the cause of this load-dependent adhesion, publishing their findings in *Nano Letters*[10]. In this manuscript, Baker et al. revealed how the adhesion force could be highly load-dependent, even while the adhesion energy remained constant.



Jacobs found this project to be a fascinating challenge. Initially, they only knew that measured work of adhesion was strongly load-dependent, and that it should not be. They talked to other experts at conferences and searched the literature. They controlled for as many factors as they could think of to make sure none were affecting the outcome.

Their findings reproduced on different materials, different loads, different dwell times and pull-off rates. The same result was obtained with the electron beam on or off (beamless tests were only possible due to the load control available with Bruker's Hysitron PI 95 TEM PicoIndenter).



After ensuring that all factors were controlled for, they were still left with the same result: the work of adhesion, measured with a conventional method, depended strongly on applied load. The only logical conclusion was that the conventional methods were not accurately describing the behavior. The next step was to carefully examine the assumptions made in those calculations of work of adhesion, and here they began to find answers. The classical models of adhesive contact describe them as a fracture problem, like a crack propagating as the two surfaces separate. It turned out that, in many cases, this is not accurate. In reality, there was a transition from fracture-controlled behavior to a “pop-off” regime where the surfaces separated suddenly without a continuous change in area prior to separation. Baker et al. were able to leverage a theoretical model of this transition to solve this experimental paradox. Correcting for this regime change and using the directly observed contact area, they were able to obtain the true work of adhesion and show that it is unchanging with load. These findings indicate that a classic technique for measuring work of adhesion, which is used in thousands of scientific publications per year, may in some cases need revision to account for the separation mechanism.

> "This work shows that even while the adhesion energy is load independent, the force to separate a nanoscale contact can increase more than seven-fold with applied load."

Summary: Breaking Down Assumptions

Typically it is not feasible to see a nanoscale contact, and so researchers must make assumptions about how that contact behaves. By directly observing and measuring nanoscale contacts using cutting-edge in-situ tools, Prof. Tevis Jacobs and his group determine where and how the conventional models fail, and attempt to devise new and more accurate descriptions of nanoscale contacts.

Recent Publications

1. Jacobs, T. D. B., and Carpick, R. W. (2013). Nanoscale wear as a stress-assisted chemical reaction. *Nature Nanotechnology*, 8(2), 108–112. https://doi.org/10.1038/nnano.2012.255
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3. Jacobs, T. D., Junge, T., and Pastewka, L. (2017). Quantitative characterization of surface topography using spectral analysis. *Surface Topography*: Metrology and Properties, 5(1), 013001. https://doi.org/10.1088/2051-672X/aa51f8
4. Vishnubhotla, S. B., Chen, R., Khanal, S. R., Martini, A., and Jacobs, T. D. (2018). Understanding contact between platinum nanocontacts at low loads: The effect of reversible plasticity. *Nanotechnology*, *30*(3), 035704. https://doi.org/10.1088/1361-6528/aaea2b
5. Vishnubhotla, S. B., Chen, R., Khanal, S. R., Li, J., Stach, E. A., Martini, A., and Jacobs, T. D. B. (2019). Quantitative measurement of contact area and electron transport across platinum nanocontacts for scanning probe microscopy and electrical nanodevices. *Nanotechnology*, *30*(4). https://doi.org/10.1088/1361-6528/aaebd6
6. Chen, R., Vishnubhotla, S. B., Jacobs, T. D. B., and Martini, A. (2019). Simulations of the effect of an oxide on contact area measurements from conductive atomic force microscopy. *Nanoscale*, *11*(3), 1029–1036. https://doi.org/10.1039/c8nr08605b
7. Milne, Z. B., Schall, J. D., Jacobs, T. D., Harrison, J. A., and Carpick, R. W. (2019). Covalent Bonding and Atomic-Level Plasticity Increase Adhesion in Silicon–Diamond Nanocontacts. *ACS applied materials and interfaces*, *11*(43), 40734-40748. https://doi.org/10.1021/acsami.9b08695
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9. Chen, R., Vishnubhotla, S. B., Khanal, S. R., Jacobs, T. D. B., and Martini, A. (2020). Quantifying the pressure-dependence of work of adhesion in silicon–diamond contacts. *Applied Physics Letters, 116*(5), 051602. https://doi.org/10.1063/1.5127533
10. Baker, A. J., Vishnubhotla, S. B., Chen, R., Martini, A., and Jacobs, T. D. B. (2022). Origin of Pressure-Dependent Adhesion in Nanoscale Contacts. *Nano Letters, 22*(14), 5954–5960. https://doi.org/10.1021/acs.nanolett.2c02016

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