SURFACE TENSION AND DEFORMATION IN SOFT ADHESION

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Abstract

In classic theories of solid adhesion, surface energy drives deformation to increase contact area while bulk elasticity opposes it. Recently, solid surface stress has been shown also to play an important role in opposing deformation of soft materials such as pressure sensitive adhesives. By studying adhesive contact between compliant gels and rigid objects, we observe that soft materials adhere very differently than their stiffer counterparts. These effects are due both to solid capillarity and to the two-phase nature of many soft solids.

Introduction

Modern contact mechanics was originally developed to account for the competition between surface energy and elasticity in adhesion with relatively stiff deformable materials, like rubber [1,2]. However, modern pressure sensitive adhesives are often orders of magnitude more compliant [3], meaning that the classic Johnson-Kendall-Roberts (JKR) [1] and Maugis [2] theories may not accurately describe their adhesive properties.

For such soft materials, the solid surface stress or surface tension can also play an important role alongside elasticity in resisting deformation. For stiff materials, any contribution from surface stress is negligibly small compared to elastic restoring forces; however, for very soft solids, the surface tension can compete with and even dominate over the elastic response [4-7]. Recently, it was shown that the JKR theory does not accurately describe adhesive contact with soft solids because it neglects the surface stress [4].

In the work described here, we explore the consequences of surface stress on adhesion between highly compliant solid gel substrates and small rigid, spherical indenters, using optical microscopy to measure the substrate deformation during contact and detachment.

Experiments

We perform experiments that bring small, rigid silica spheres into adhesive contact with compliant silicone substrates. There are two experimental configurations: with zero applied external force, in which the spheres are placed on the sticky substrates and spontaneously indent to an equilibrium position [4,7], and with an applied external load, in which we manipulate the position of the sphere in order to make and break contact with the substrate.

The spheres range in radius from 7 to 35 micrometers. The surface of the spheres is either bare silica or surface-functionalized with a hydrocarbon silane, which lowers the adhesion energy.

The adhesive substrates are lightly crosslinked, solid, polydimethylsiloxane (PDMS) gels with a Young modulus of 5.6 kPa and a Poisson ratio of 0.48. The fraction of free liquid PDMS in these gels is measured by solvent extraction in toluene to be 62%.

We image the sphere and the deformed silicone substrate using optical microscopy. Confocal microscopy allows us to measure the conformation of the PDMS substrate around the indenting spheres in 3D, while brightfield microscopy of the deformation profile allows us to measure the sphere position and long-range substrate deformation to very high precision. Brightfield snapshots from an example pull-off experiment are shown in Figure 1 *(top)*.

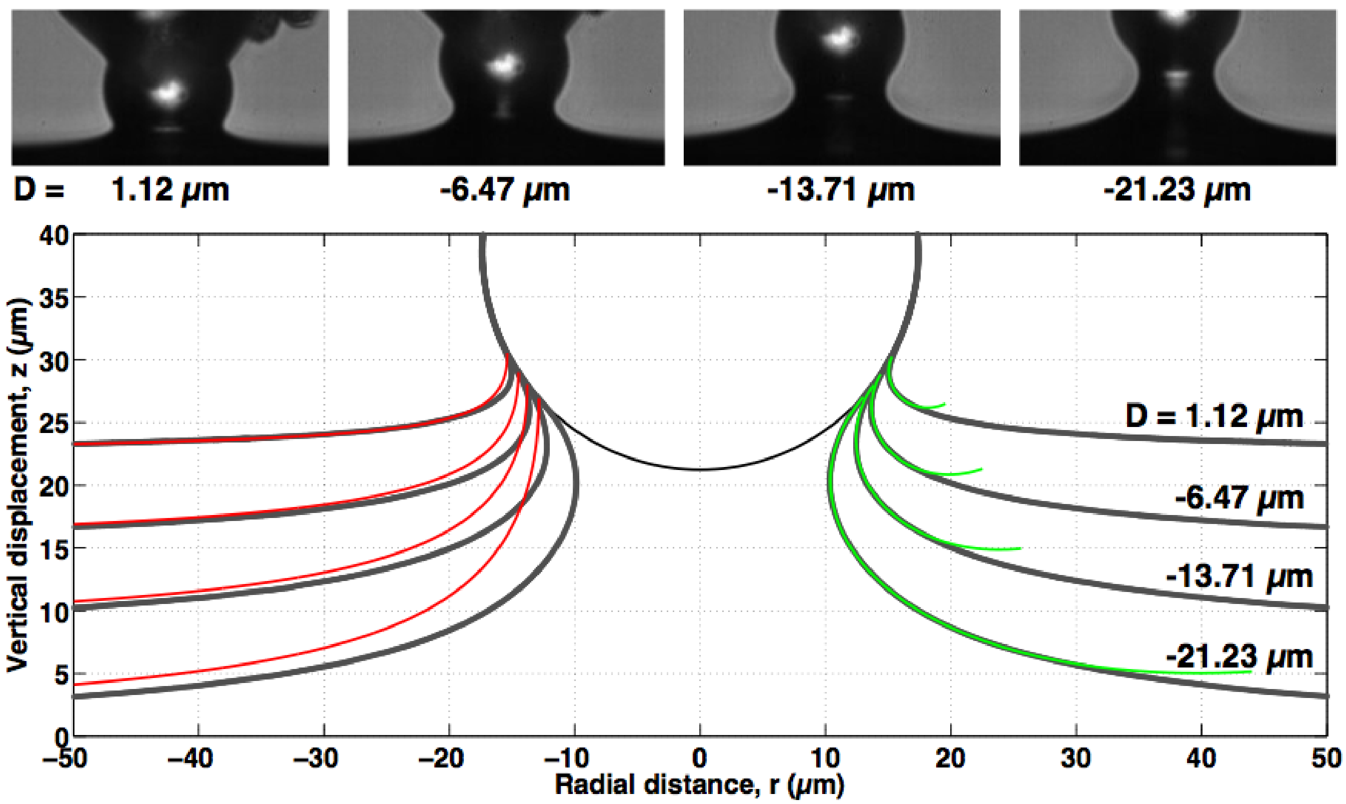

Figure 1. A rigid spherical indenter brought into adhesive contact with a compliant PDMS substrate, then pulled away. *(top)* Snapshots of a 5.6kPa silicone substrate deforming as it maintains stable adhesive contact with a 17-μm-radius rigid silica sphere during a pull-off experiment. *(bottom)* Mapped profiles of the sphere and silicone surface (gray) at positions corresponding to the above snapshots, shifted so that the sphere position is constant. Superimposed curves show the best-fit elastic theory (red, left) [2] and capillary solutions [8] (right, green).

Results

From raw images such as those shown in Figure 1 *(top)*, we map the 3D deformation profile of the PDMS surface to ~10 nm resolution as we slowly pull the sphere away from contact. These high-resolution profiles allow us to measure the sphere position and indentation depth, ***D***, defined with respect to the original, undeformed surface of the silicone gel substrate. We show the mapped profiles for four positions during an example touch-and-pull experiment as gray points in Figure 1 *(bottom)*. In order to emphasize the substrate deformation, these profiles are shifted so that the sphere position remains constant (black circle). All of these are stable deformation configurations; adhesive detachment has not begun.

We compare our measured deformation profiles to the predictions of the classic elastic theory [1,2]. These predictions are superimposed as red lines on the left side of Figure 1 *(bottom)*. We observe that the classic theories describe the deformation of the substrate fairly well far from the indenter, but completely fail close to the contact line. In particular, they never capture the "solid meniscus" shape of the deformed surface, nor do they accurately predict the observed zero-degree contact angle.

By contrast, on the right side of Figure 1 *(bottom)* we show the results of fitting the data close to the contact line with a surface of constant total curvature, superimposed as green lines on the Figure. This is the shape that would result from capillary-dominated mechanics [8], in which surface stress completely overwhelms elasticity. We find that this capillary solution describes the deformation of the adhesive substrate extremely well in the near field close to the contact line, but fails far from the contact line. Surprisingly, the domain of capillary dominance increases as the sphere is pulled farther and farther from its initial contact.

Conclusions

The similarities—and differences—between our experimental measurements and the classic theories point to a crossover from a capillary-dominated near field response close to the contact line to an elastic-dominated response in the far field. Surprisingly, we find that the profile shape is increasingly capillary-dominated with increasing displacement, pointing to unexpectedly far-reaching surface tension effects during adhesive contact with applied force.

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