Stretchable gold conductors on elastomeric substrates

Stéphanie Périchon Lacour[a)] and Sigurd Wagner
Department of Electrical Engineering, Princeton University, Princeton New Jersey 08544

Zhenyu Huang and Z. Suo
Department of Mechanical and Aerospace Engineering and Princeton Material Institute, Princeton University, Princeton New Jersey 08544

(Received 27 December 2002; accepted 13 February 2003)

Stripes of thin gold films are made on an elastomeric substrate with built-in compressive stress to form surface waves. Because these waves can be stretched flat they function as elastic electrical conductors. Surprisingly, we observe electrical continuity not only up to an external strain of ~2% reached by stretching the films first flat (~0.4%) and then to the fracture strain of free-standing gold films (~1%), but up to ~22%. Such large strains will permit making stretchable electric conductors that will be essential to three-dimensional electronic circuits. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1565683]

Many large-area electronic circuits will need electrical conductors that can sustain large mechanical strain, either once or reversibly. Retina-shaped photosensor arrays[1] must undergo a one-time large deformation from flat to spherical, and connectors to electroactive polymers,[2,3] stretchable sensor skin,[4] and electronic textiles[5] all may rely on stretching. One-time deformation may be accommodated by plastic flow of the metal or of a sacrificial material.[1] Electrical interconnects that can be stretched and relaxed reversibly are also needed and require new design concepts. Typical free-standing metal films fracture at tensile strains below ~1%.[6] It has been observed that when metal films are expanded during the deformation of a substrate they fracture at strains of a few percent.[7]

We made stretchable interconnects by exploiting the compressive stress that can be built into thin films of gold evaporated onto elastomeric membranes of polydimethyl siloxane (PDMS). Compressive stress in the gold films induces spontaneous wrinkling,[8] which can shrink the net length of the thin-film conductors by several tenths of a percent. Intending to make use of this strain to raise the stretchability of the gold films above their fracture strain of, typically, ≈1%, we set out to study spontaneously wrinkled gold stripes for use as elastic interconnects. In the course of this study we discovered that the stripes can retain electrical continuity when stretched by as much as 22%.

Thin metal films—typically, 100-nm-thick layers of gold on top of a 5-nm-thick adhesion interlayer of chromium—were deposited in one run by successive electron beam evaporation onto elastomeric substrates of PDMS held at room temperature. We chose gold as one of the most ductile metals and with high electrical conductivity. 1-mm-thick substrates of PDMS (Sylgard 184® by Dow Corning) were prepared by mixing the silicone gel with cross linker in a 10:1 weight ratio, spreading it, and curing for at least 12 h at 60 °C. PDMS, so prepared, is compatible with microelectronic clean room equipment.

When the metal film extends over a large surface area, random wrinkles cover the sample except at the film's edges, as shown in Fig. 1(a). When the metal is deposited through a shadow mask to define stripes, surface waves form as shown for the 0.25-mm-wide stripe of Fig. 1(b). We now proceed to discuss the mechanics of buckling and wave formation, and then describe our subsequent discovery of very large stretching with retention of electrical conductance.

The thin metal film is deposited with compressive stress. The compressive stress causes the film to buckle.[8] Complex wave structures of thin films on compliant substrates have been studied by several groups.[8–11] Large compressive stresses can be introduced by prestretching the substrate.[12] To model the buckling of our films we relate the stress within the thin metal film/PDMS structure to the wave pattern. We made the following assumptions: the metal film is ideally bonded to the rubber substrate; the PDMS substrate is incompressible, i.e., its Poisson ratio $\nu_{PDMS}=0.5$; its thickness is infinite compared to that of the Au layer; shear at the metal/PDMS interface is neglected; and the film has thickness h. The as-deposited compressive strain in the film ε_0 is produced by built-in strain and thermal expansion mismatch ($\alpha_{Au}=14\times10^{-6}$/K, $\alpha_{PDMS}=960\times10^{-6}$/K). The film can relax in both the x and y directions in response to its initial compressive strain. We use nonlinear perturbation and energy minimization methods[13] to determine the film's critical strain ε_c (the minimum compressive strain at which it

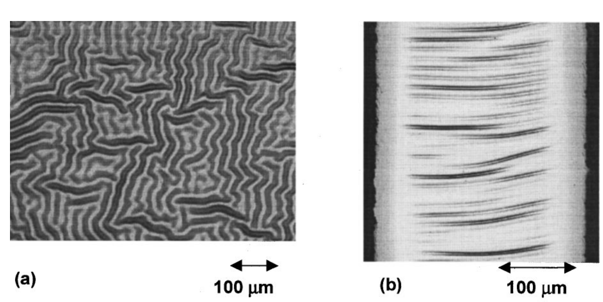

FIG. 1. Optical images of the wave pattern formed in a 100-nm-thick gold film evaporated on a 1-mm-thick PDMS membrane (a) over the entire surface; (b) of a stripe evaporated through a shadow mask. Dark margins are from the PDMS substrate.

[a)]Electronic mail: slacour@princeton.edu

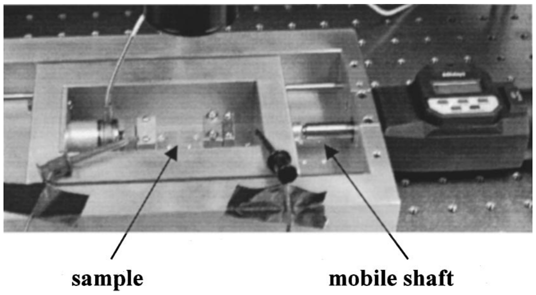

FIG. 2. Photograph of the custom made microtensile tester.

buckles—it will stay flat if $|\varepsilon_0|<|\varepsilon_c|$), the equilibrium buckling wavelength λ, and its amplitude A. When stretched, the film becomes flat at $\varepsilon_{\mathrm{external}}=\varepsilon_c-\varepsilon_0$.

For a 100-nm-thick Au layer (Young's modulus 82 GPa), and with $E_{\mathrm{PDMS}}=1.2$ MPa measured under tensile stress, the predicted wavelength is 21 μm, while we measured 10 ± 1.5 μm with the atomic force microscope in the tapping mode. The measured peak-to-valley amplitude is 0.45 ± 0.15 μm. The calculated critical strain for buckling ε_c is 0.016%.

When we define conductor stripes, we evaporate Cr/Au through a shadow mask made of polyimide foil. When the polyimide foil is taken off, the stress in the metal film is released and no longer is equiaxial. The structure relaxes laterally at the edges of the gold lines, and a pattern of parallel waves develops with its crests aligned perpendicularly to the direction of the maximum compressive stress as shown in Fig. 1(b). This pattern is a regular sinusoid with a wavelength similar to that of the random buckling.

We evaluated the electrical resistance of the wavy Au lines while under mechanical strain. Electrical contacts of conducting epoxy paste were applied, and 0.1-mm-diam gold wires were embedded in the paste. The paste burying the wires was sandwiched between the gold film/PDMS substrate and a second piece of PDMS to ensure electrical connection as well as mechanical compliance. A sample is shown in Fig. 2 mounted under a microscope in a strain tester and with electrical leads. The Au stripe is 100 nm thick, 28 mm long, and 0.25 mm wide. The electrical resistance as a function of uniaxial strain was measured *in situ* using a Keithley 4210 source meter. The lengthening of the sample and eventual development of cracks in the Au stripe were recorded with an Infinity long distance K2 microscope, a Kodak digital camera, and an *XYZ* translation platform. The elongation was measured with a 1-μm-resolution Mitutoyo meter. From this elongation $L-L_0$, the values of externally applied strain $\varepsilon_{\mathrm{external}}=(L-L_0)/L_0$ are calculated from the difference of the sample length, L, to its unstressed length, L_0. We began by measuring the electrical resistance without external strain. Then, the sample was elongated, first in 0.18% steps of $\varepsilon_{\mathrm{ext}}$, and beyond $\varepsilon_{\mathrm{ext}}=6\%$ in 0.36% steps, and was held for 5 min at each strain value. Figure 3 presents the change in electrical resistance $(R-R_0)/R_0$ normalized to its initial value, with the tensile strain.

After mounting in the test frame and in the absence of externally applied strain, the Au stripe of Fig. 3 was buckled, as shown in Fig. 1(b). The surface of the sample became flat after the first two 0.18% steps$=$0.36% external strain were

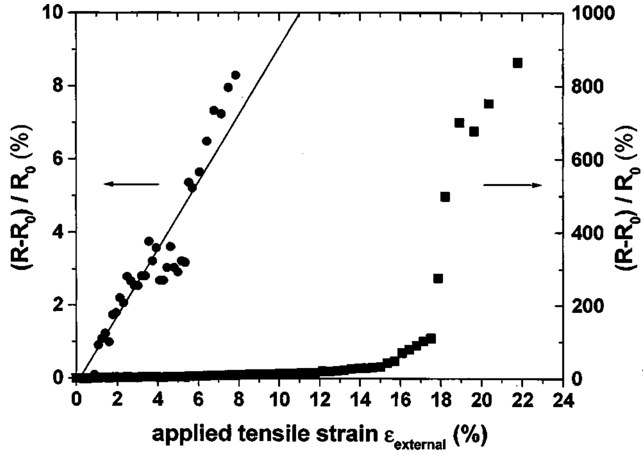

FIG. 3. Variation of the normalized change in electrical resistance $(R-R_0)/R_0$ of a 100-nm-thick Au stripe with applied tensile strain $\varepsilon_{\mathrm{external}}$ (right curve). The left curve represents the linear behavior of $(R-R_0)/R_0$ with the applied external strain for $0<\varepsilon_{\mathrm{external}}<8\%$.

applied; it was free of cracks. Because the calculated critical strain ε_c is only 0.015%, the initial compressive strain in the gold film ε_0 is near 0.36%.

We then went on to measure resistance under tension, expecting to reach a maximum value of tensile strain in the electrically continuous film of $\sim 1\%$. Figure 3 shows that, surprisingly, the Au line remains conducting under $\varepsilon_{\mathrm{external}}$ much above this typical fracture strain of a free-standing thin film. Figure 3 exhibits two regimes. At strains lower than $\sim 8\%$, $(R-R_0)/R_0$ is proportional to elongation. In this regime small cracks appear at the edges of the stripe, as shown in Fig. 4(a). At higher strain, cracks extend across much of the width of the stripe, perpendicular to its long dimension and visible in Fig. 4(b). These longer cracks cause a pronounced rise of the electrical resistance. At strains above 15%, these cracks traverse the full width of the Au line. However, while the electrical resistance rises drastically, it remains finite. This observation suggests that the film is bro-

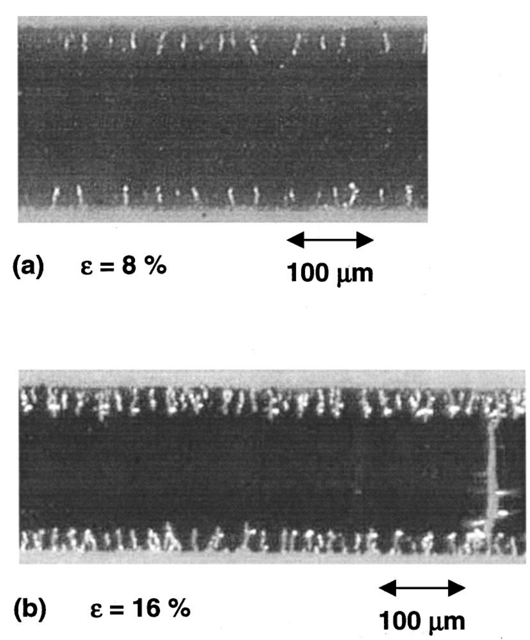

FIG. 4. *In situ* photographs of a 100-nm-thick Au stripe on 1-mm-thick PDMS under (a) 8% and (b) 16.4% tensile strain.

ken, but that a thin conductive layer remains at the bottom of the cracks.[6,14] From the crack dimensions we estimate that this conducting remnant layer is one metal atom thick. We have not yet been able to detect the material of this layer. At $\varepsilon_{\mathrm{external}}=23\%$ the resistance became infinite.

In summary, additional stretchability of a few tenths of a percent is obtained by inducing a surface wave in gold stripes. Under tensile strain, the buckling disappears when the external strain matches the as-deposited compressive strain in the film (minus the small critical strain for buckling), which allowed us to experimentally estimate its value. We discovered that a metal film on an elastomer substrate can be stretched much beyond the fracture strain of a free-standing metal film of, typically, 1%, and remain electrically continous. We ascribe this large strain to a distribution of the externally applied strain over the entire length of the stripe. This behavior provides a path to making electrical conductors that can be stretched by about 10%. By contrast, free-standing thin metal films and whiskers, subject to small overall tensile strains, rupture by localized plastic deformation, such as local thinning or forming shear bands.[15–17] For a metal film well bonded on a compliant substrate, such localized deformation is suppressed at small strains. Of course, if the film and the substrate debond over a length much larger than the film thickness, the film becomes free standing, and will rupture by localized deformation under small overall tensile strains.

The authors gratefully acknowledge the help of Professor Dan Mumm in building the stretching apparatus, and thank Professor Tony Evans for granting its use. This research is supported by the National Science Foundation, DARPA, and the New Jersey Commission on Science and Technology.

[1] P.-H. Hsu, R. Bhattacharya, H. Gleskova, Z. Xi, Z. Suo, S. Wagner, and J. C. Sturm, Appl. Phys. Lett. **81**, 1723 (2002).
[2] M. Benslimane, P. Gravesen, and P. Sommer-Larsen, Proc. SPIE **4695**, 150 (2002).
[3] G. Kofod, R. Kornbluh, R. Pelrine, P. Sommer-Larsen, and R. Heydt, Proc. SPIE **4329**, 141 (2001).
[4] V. J. Lumelsky, M. S. Shur, and S. Wagner, IEEE Sensors J. **1**, 41 (2001).
[5] S. Wagner, E. Bonderover, W. B. Jordan, and J. C. Sturm, Int. J. High Speed Electron. Syst. **12**, 1 (2002).
[6] H. Huang and F. Spaepen, Acta Mater. **48**, 3261 (2000).
[7] P.-H Hsu, M. Huang, S. Wagner, Z. Suo, and J. C. Sturm, Mater. Res. Soc. Symp. Proc. **621**, Q8.6.1 (2000).
[8] N. Bowden, S. Brittain, A. G. Evans, J. W. Hutchinson, and G. W. Whitesides, Nature (London) **393**, 146 (1998).
[9] N. Bowden, W. T. S. Huck, K. E. Paul, and G. M. Whitesides, Appl. Phys. Lett. **75**, 2557 (1999).
[10] J. Kim and H. H. Lee, J. Polym. Sci., Part B: Polym. Phys. **39**, 1122 (2001).
[11] R. Huang and Z. Suo, J. Appl. Phys. **91**, 1135 (2002).
[12] M. Watanabe, H. Shirai, and T. Hirai, J. Appl. Phys. **92**, 4631 (2002).
[13] S. P. Timoshenko and J. M. Gere, *Theory of Elastic Stability* (McGraw-Hill, New York, 1961), pp. 319–411.
[14] D. R. Cairns, R. P. Witte II, D. K. Sparacin, S. M. Sachsman, D. C. Paine, G. P. Crawford, and R. R. Newton, Appl. Phys. Lett. **76**, 1425 (2000).
[15] C. F. Elam, Proc. R. Soc. London, Ser. A **115**, 133 (1927).
[16] D. W. Pashley, Proc. R. Soc. London, Ser. A **255**, 218 (1960).
[17] S. S. Brenner, in *Growth and Perfection of Crystals*, edited by R. H. Doremus, B. W. Roberts, and D. Turnbull (Wiley, New York, 1958), pp. 157–190.