

Stiff subcircuit islands of diamondlike carbon for stretchable electronics

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Stretchable electronics on elastomeric substrates requires fragile and brittle device materials to be placed on stiff, mechanical distinct subcircuit islands. We deposited a diamondlike carbon (DLC) film at room temperature on a silicone substrate by pulsed laser ablation, and patterned the film into an array of $200 \times 200 \mu\text{m}^2$ islands. When the substrate was uniaxially stretched to a strain of 25%, the islands remained adherent to the substrate and only deformed by $\sim 5\%$, while the exposed substrate stretched by more than 30%. A row of 11 DLC islands interconnected with gold stretchable metallization maintained end-to-end electrical conduction during a mechanical cycle to 20% tensile strain. This demonstration of electrically interconnected stiff islands on silicone illustrates two important steps toward fully integrated, elastically stretchable electronics. © 2006 American Institute of Physics. [DOI: 10.1063/1.2210170]

I. INTRODUCTION

Stretchable electronics is pushing the limits of large-area integrated circuit technologies to make electronic systems with radically different performance and functionality than conventional silicon based devices.^{1–5} Such elastic systems will be built on elastomeric substrates, which can be made to conform to any surface shape, once or many times. Applications include large-area sensor arrays for nondestructive health monitoring, electrotexiles, and smart prostheses. Applications in biomedical engineering will be particularly attractive as biocompatible stretchable electronics will enable the development of electrically and mechanically matched interfaces between machines and living tissues.⁶

A main challenge to implement stretchable electronics is how to integrate stiff and fragile device materials such as silicon with compliant elastomeric substrates such as silicones. Furthermore, today's electronic industry, whether on semiconductor substrates or large-area glass plates, is based on a planar technology, and has been optimized for stiff and flat formats. Our approach toward the fabrication of stretchable electronics has been to fabricate devices on elastomeric substrates using conventional planar technology, and to design an architecture that prevents fragile materials from fracture while enabling full elasticity of the finished system.

This paper describes one basic component, stiff subcircuit islands that mechanically isolate active devices from the stretchable substrate. We first review the architecture of stretchable electronics. Then we describe the preparation of diamondlike carbon subcircuit islands on a silicone substrate and their interconnection with stretchable gold metallization. We conclude with experiments on islands of silicon oxide and silicon nitride.

II. ARCHITECTURE OF STRETCHABLE ELECTRONICS

Current flexible electronics focuses on cylindrical deformation produced by bending or rolling.⁷ The ensuing strain in device materials can be minimized by placing them in the neutral plane of the structure.⁸ With proper design, the maximum strain can be kept below a few tenths of a percent. The applications envisioned for stretchable electronics require deformation by tens of percents.

In previous work, we have shown that a metallic thin film, i.e., a *ductile* material can elongate substantially without rupture if well bonded to a compliant substrate.^{9,10} The adherent substrate distributes the strain of incipient necks in the metal and thereby suppresses necking and rupture. Additionally, twisting and bending out of the plane of the metal film on elastic substrate also allows for large elongation.¹¹ On the other hand, most other electronic materials, such as silicon and silicon nitride, are *brittle*. They crack at strains of a few tenths of percent.¹² Therefore, it is essential to reduce the strains in the brittle materials. Our approach has been to pattern brittle materials onto stiff islands. Such an approach has been used to fabricate electronic circuits on polymer foil, which was then plastically deformed to a dome.^{13–15}

Figure 1 sketches the architecture of stretchable electronics. The substrate is typically a polydimethylsiloxane (PDMS) membrane of thickness $h_{\text{sub}} \sim 1$ mm and Young's modulus $E_{\text{sub}} \sim 1$ MPa. Patterned on the substrate are subcircuit islands of a stiff material, on which devices are fabricated; the combined thickness of the island and the devices may be as small as $1 \mu\text{m}$. Stretchable metallization electrically connects neighboring subcircuits. When the structure is deformed by a large strain, the islands deform by small strains and remain intact, while elongation and compression are distributed in the substrate and interconnects.

Cross sections of the island structure are shown Fig. 2. Assume that the array is made of a series of n identical cells

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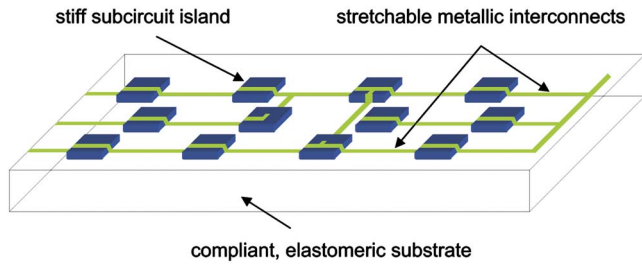


FIG. 1. (Color online) Architecture of elastic electronics. A compliant, elastomeric substrate carries stiff subcircuit islands interconnected with stretchable metallization. Upon deformation, the substrate along with the stretchable interconnects accommodates the resulting tensile, compressive, and shear strains while the islands remain intact.

composed of an island of side a and an interisland spacing of length b . Also assume that the island stays rigid under stretching, i.e., is not deformable, and that the interisland distance becomes $b + \Delta b$ upon stretching. The applied strain ϵ_{appl} distributes as

$$\epsilon_{\text{appl}} = \frac{n(a + b + \Delta b) - n(a + b)}{n(a + b)} = \frac{\Delta b}{a + b}. \quad (1)$$

One can then relate the interisland strains $\epsilon_{\text{inter},x}$ and $\epsilon_{\text{inter},y}$ to the applied strain using the following equation:

$$\epsilon_{\text{inter},x} = \frac{n\Delta b}{nb} = \left(1 + \frac{a}{b}\right) \epsilon_{\text{appl}}, \quad (2)$$

$$\epsilon_{\text{inter},y} = -\nu_{\text{sub}} \epsilon_{\text{inter},x} = -\nu_{\text{sub}} \left(1 + \frac{a}{b}\right) \epsilon_{\text{appl}}, \quad (3)$$

with ν_{sub} the Poisson ratio of the substrate, which is 0.5 for an elastomer.

To guarantee the architecture function, appropriate island material, size, and density must be chosen, and electrical continuity of the interconnects running off the island edges must be ensured. The island material should preferably be an electrical insulator that can be deposited with thin-film techniques at low substrate temperature, to be compatible with the organic substrate. The stiffness S of a layer of a material with thickness h and Young's modulus E is characterized by

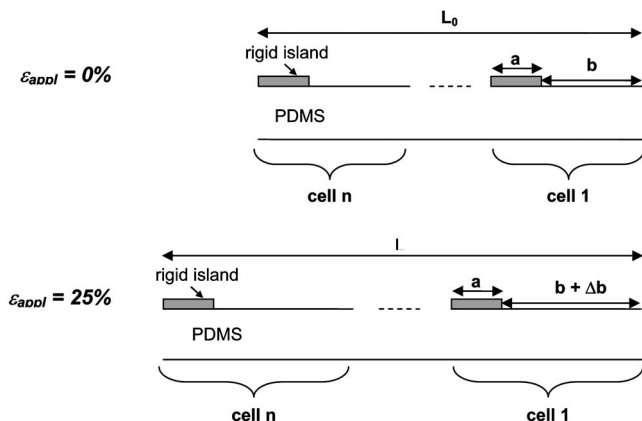


FIG. 2. Cross-sectional sketches of a rigid island array. The array consists of n identical cells composed of a rigid island of side a and an interisland distance b . Upon uniaxial stretching, the interisland distance increases to $b + \Delta b$. The rigid island remains undeformed.

the product $S = Eh$. Assuming $h_{\text{sub}} = 1$ mm and $h_{\text{film}} = 250$ nm, for the film to be stiffer than the substrate, the film should have a Young's modulus at least 4 000 times that of PDMS, i.e., $E_{\text{film}} > 4$ GPa. Candidates for the island material include silicon oxide ($E_{\text{SiO}_x} \sim 70$ GPa), silicon nitride ($E_{\text{SiN}_x} \sim 200$ GPa), and diamondlike carbon ($E_{\text{DLC}} > 200$ GPa). The DLC films have recently attracted interests for tribological, optical, electronic, and biomedical applications because of their high hardness, wear resistance, chemical inertness, and an optical gap that can be varied from zero to a few eV.^{16–18} Finally, island size and density should be optimized to accommodate built-in transistor circuits and prevent crack propagation upon stretching. Typically islands larger than the substrate thickness crack because of shear stress pulling on the island during stretching.^{13,15} Also high island density reduces the yield of intact island during deformation.¹⁵

Using the finite element method (FEM), we simulated the deformation of a SiN_x island of radius $250 \mu\text{m}$ on a PDMS substrate subject to elongation. The island/substrate thickness ratio was 10^{-3} , which would correspond to a $1 \mu\text{m}$ thick island on a 1 mm thick substrate. The structure was assumed to be cylindrically symmetric with a constant displacement load applied on the outer surface of the substrate. The simulation showed that the strain within the island is negligible, while the elongation is mainly accommodated by the substrate. The simulation also revealed the strain concentration in the substrate near the island edge. While the strain in the island edge remains insignificant, the surrounding substrate may stretch by several tens of percent to accommodate the overall elongation. This is important as interconnects running off the island edges will thus have to withstand large local strain. We shall show that a gold interconnect can withstand this strain without electrical failure.

III. ISLAND PREPARATION AND CHARACTERIZATION

A. Diamondlike carbon

We used polydimethylsiloxane prepolymer (Sylgard® 184, Dow Corning) to prepare 1 mm thick elastomeric membranes. After mixing the prepolymer with its crosslinker in a 10:1 weight ratio, the blend was cast in plastic petri dishes and cured for at least 24 h in an oven at 60°C . Once released from the dishes, the PDMS membranes were mounted on plastic backing foils for easy handling.

We describe in details the most successful island material, diamondlike carbon film, and in more cursory form the two other island materials, SiO_x and SiN_x .

The DLC films were deposited on the PDMS substrate by pulsed laser ablation. A KrF excimer laser ($\lambda = 248$ nm) was used to ablate carbon species from a high purity graphite target, with the substrate close to room temperature,^{16,18} which minimizes thermal stress in the DLC film caused by the large difference in the coefficients of thermal expansion of the PDMS ($\text{CTE}_{\text{PDMS}} = 300 \text{ ppm}/^\circ\text{C}$) and DLC ($\text{CTE}_{\text{DLC}} \sim 2 \text{ ppm}/^\circ\text{C}$). The deposition rate was $\sim 6 \text{ nm}/\text{min}$. DLC islands were 185 nm thick.

Stretchable metallization was prepared by electron-beam evaporation.² A thin layer of chromium was first deposited on the elastomeric substrate as an adhesion layer, followed

by a 25 nm thick subsequent gold film. The maximum temperature reached at the end of the evaporation was 60 °C.

B. Patterning of DLC islands and interconnects

The islands and the interconnects were patterned by lift-off and shadow masking.^{2,19,20} We avoid using spinnable photoresist because PDMS swells in the solvents employed in photolithography.²¹

Large DLC patches of side $a > 1$ mm were prepared using a Kapton shadow mask. 100 μm to 1 mm DLC islands were patterned with Riston dry photoresist (Riston®, DuPont), which after UV exposure and development were laminated on the bare PDMS surface. After DLC deposition, the Riston was stripped off in a KOH solution for ~ 20 min. The samples were then soaked in de-ionized water for about the same time and blow dried with a nitrogen gun.

Interconnects were also patterned using a Kapton shadow mask. Openings for ~ 2 cm long, several hundred micron wide stripes were made in a Kapton foil that was subsequently stuck on the PDMS substrate. After metal evaporation, the mask was carefully peeled off to avoid damaging the metallization.

C. Characterization of DLC islands and interconnects

All samples were observed with an Olympus SX9 stereomicroscope and an Olympus MX40F optical microscope with a digital camera. The surface microstructure was characterized using a Philips XL30 scanning electron microscope (SEM) after sputtering a 1.5 nm thick layer of iridium onto each sample.

To evaluate their stretchability, island samples were mounted between two clamps in a uniaxial stretcher and stretched manually using a 1 μm resolution Mitutoyo meter.¹⁹ Upon stretching, one clamp remained fixed, while the other clamp was pulled away. The stretching direction was parallel to the length of the sample, along the x axis. The applied strain $\epsilon_{\text{appl}} = \Delta L / L_0$ was calculated from the measured elongation ΔL of the sample and its initial length L_0 . The samples were cycled between $\epsilon_{\text{appl}} = 0\%$ and 25% in strain increments/decrements of 2%–5%. Island dimensions as well as distances in between the islands were measured with an accuracy of ± 5 μm along the x (stretching direction) and y axes from micrographs taken during stretching cycles. For each strain increment, dimensions were averaged over 2–10 islands. From each average, we calculated the strains in the islands (ϵ_x and ϵ_y) and in between the islands ($\epsilon_{\text{inter},x}$ and $\epsilon_{\text{inter},y}$).

The electromechanical characteristics of gold interconnects running over several subcircuit islands were recorded in the same uniaxial stretcher but driven by a stepper motor.²² The sample was stretched to $\epsilon_{\text{appl}} = 20\%$ with strain increments/decrements of 0.5% strain every 5 min. The electrical resistance of the gold interconnect was measured with a Keithley 4140 sourcemeter.

D. Silicon oxide and silicon nitride

Arrays of 400 nm thick SiO_x islands were prepared by electron-beam evaporation on PDMS substrate, and pat-

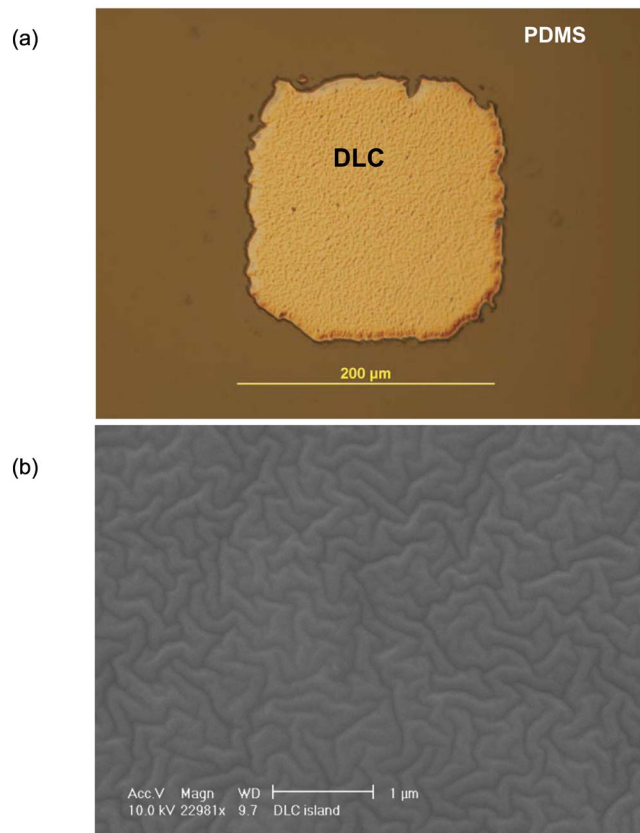


FIG. 3. (Color online) (a) Optical micrograph of a $200 \times 200 \mu\text{m}^2$, 185 nm thick DLC island on 1 mm thick PDMS substrate. (b) SEM picture of the wrinkled DLC surface. The wrinkle wavelength is ~ 175 nm.

terned by lift-off using Riston dry photoresist. The islands were squares with $a = 100 \mu\text{m}$ side and 100 μm apart.

Silicon nitride islands were prepared by plasma enhanced chemical vapor deposition (PECVD) at 150 °C substrate temperature. 5×5 arrays of 2 mm diameter, ~ 300 nm thick SiN_x islands were prepared through a stainless steel shadow mask. SiN_x islands were spaced 4 mm apart.

IV. STRETCHING OF ARRAYS OF DLC ISLANDS

Before mechanical deformation, all samples were optically inspected. DLC films adhered well to the PDMS substrate; no delamination was observed at the edges of the islands. As DLC films are known to adhere well to silicon substrates,¹⁷ the silicone surface may favor DLC adhesion. Islands larger than 1 mm cracked either immediately after DLC deposition or after mask removal. Islands smaller than 1 mm were crack-free. Figure 3(a) shows a 200 μm DLC island on PDMS. Lifting-off the Riston photoresist left the island edges ragged, and the contour of the initial $200 \times 200 \mu\text{m}^2$, which opened in the Riston film, is still inscribed in the PDMS. Under the SEM, we observed a wrinkled DLC surface with a wavelength of ~ 175 nm [Fig. 3(b)]. The wrinkling results from the large compressive internal stress in the DLC films built during the growth.²³

Figure 4(a) shows a 17×17 array of $a = 200 \mu\text{m}$ islands; neighboring islands are 400 μm apart. The sample is seen to be clamped in the uniaxial stretcher. The (initial) distance L_0 between the clamps was 11 mm. Figure 3(b) shows the

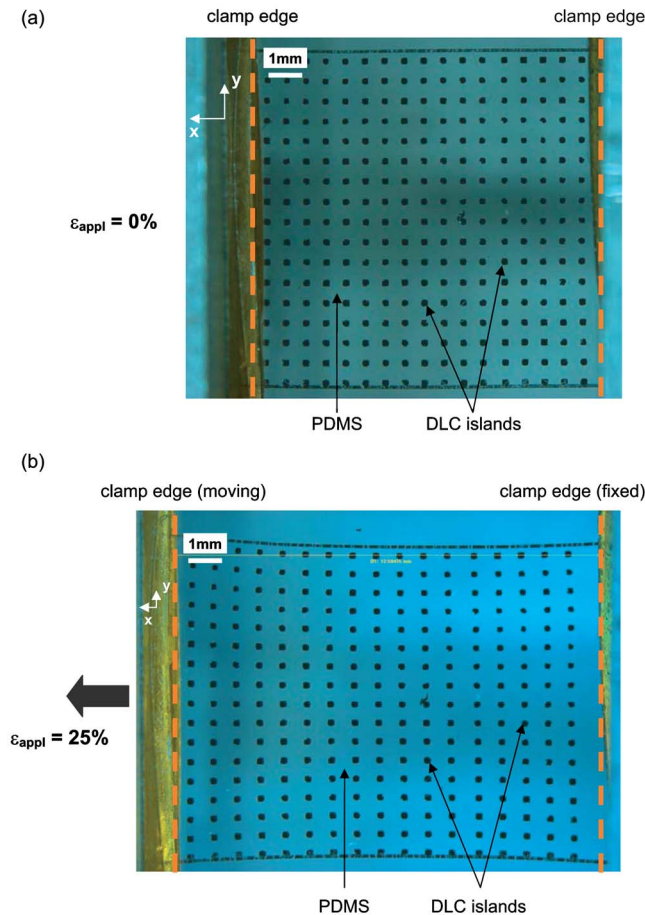


FIG. 4. (Color online) Top view of a 17×17 array of a $200 \mu\text{m}$ side DLC islands on PDMS substrate: (a) before stretching; (b) while stretched along the x axis by 25% applied tensile strain.

sample at maximum elongation, $\epsilon_{\text{appl}} = 25\%$. The maximum lateral contraction of the elastomeric substrate, which is clearly visible along the y axis, was $\sim -8.7\%$. None of the DLC islands cracked during stretching. The dimensions of the islands changed far less than the spacing between the islands, which increased along the x axis and decreased along the y axis. Figure 5 plots the strains along both x and y axes within the islands and in between the neighboring is-

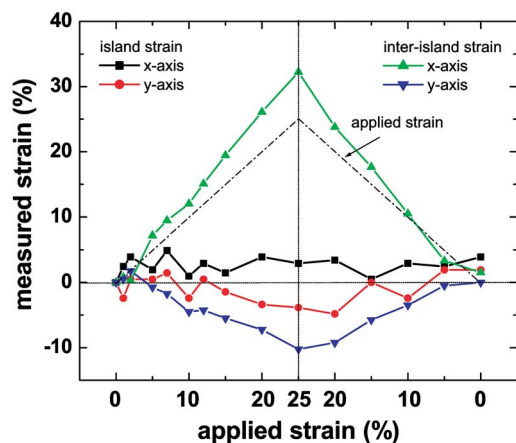


FIG. 5. (Color online) Measured strains in DLC islands and in between the islands as a function of the applied strain.

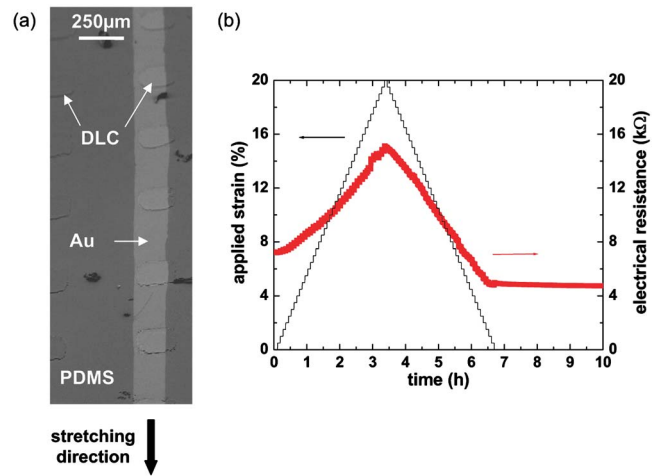


FIG. 6. (Color online) (a) SEM picture of a portion of the $\sim 200 \mu\text{m}$ wide gold interconnect after one stretching cycle. (b) Electrical resistance of a gold interconnect running across 11 DLC islands as a function of time and applied mechanical strain.

lands for an applied (machine) strain ϵ_{appl} that raised from 0% to 25%, and then returned to 0%. The islands strained to a maximum of 5% did not break but stretched elastically. Previous work by Ogwu *et al.* showed that Si-DLC coatings on stainless steel substrates can stretch to $\sim 5\%$ tensile strain without fracture.²⁴ On the other hand, the strain between the islands was large. The interisland strain, $\epsilon_{\text{inter},x}$ ($\epsilon_{\text{inter},y}$) is seen to rise with ϵ_{appl} to a maximum $\epsilon_{\text{inter},x} \sim 35\%$ in the x direction and of $\epsilon_{\text{inter},y} \sim -11\%$ in the y direction. The strains in the structure nearly vanished when fully unloaded, indicating that the structure is almost elastic.

The observed interisland strains agree with the calculation of $\epsilon_{\text{inter},x}$ and $\epsilon_{\text{inter},y}$ using Eqs. (1) and (2). For an a/b ratio of 0.5, Eq. (2) predicts $\epsilon_{\text{inter},x} \sim 37.5\%$ and $\epsilon_{\text{inter},y} \sim -18.75\%$. Calculated and experimental tensile strains are of the same order. But the measured compressive strain $\epsilon_{\text{inter},y}$ is smaller than the theoretical one. This may result from a limited “Poisson’s compression” of the sample held between the two wide clamps (shown in Fig. 4).

$200 \mu\text{m}$ side DLC islands on PDMS substrate are stiff, and can stand up to stretching without fracture. Given our DLC array pattern ($a/b=0.5$), DLC islands remained intact over stretching cycle to 25%, but deformed by $\sim 5\%$, which is still greater than the fracture strain of device materials. One way to maintain island strain below 1% would be to reduce further the island size and increase the interisland spacing.

V. STRETCHING OF ELECTRICALLY INTERCONNECTED DLC ISLAND ARRAYS

While the strain in the islands must remain below the critical fracture strain of device materials, the interconnects must strain fully with the substrate and remain electrically conducting. Therefore, we evaluated the stretchability and electrical continuity of metallic interconnects running over several DLC islands. Figure 6(a) presents a SEM picture of a portion of a gold interconnect patterned over several DLC islands. The width of the gold stripe covers most of the DLC islands, and a small portion of the gold continues on the bare

PDMS. This picture was taken after relaxation from a stretching cycle to a strain of 20% along the length of the interconnect. The DLC islands and the Au film are completely intact.

A test gold conductor, running only on bare PDMS, was prepared on the same PDMS substrate. The gold film electrical resistivity was found to be $\sim 5.10^{-4} \Omega \text{ cm}$. Au films on PDMS are typically 2–50 times more resistive than Au film on glass.²⁵ This particular batch of samples was almost 100 times more resistive than Au film on glass. The initial resistance of the 200 μm wide, 1 cm long interconnect shown in Fig. 5(a) was $\sim 7.2 \text{ k}\Omega$, in agreement with the measured gold resistivity.

Figure 6(b) presents a test run in which the gold interconnect running over 11 DLC islands was subjected to a stretching cycle up to $\epsilon_{\text{appl}}=20\%$ and back to 0%. The electrical resistance increased (decreased) almost linearly with the applied strain. At $\epsilon_{\text{appl}}=20\%$, $R_{\text{max}} \sim 15 \text{ k}\Omega$. Despite the large applied strain and the tortuous path over several island steps, the gold interconnect remained electrically conductive. After the stretching cycle, its electrical resistance relaxed to $R_f \sim 4.7 \text{ k}\Omega$. Optical micrographs of the gold film covering a DLC island before, during, and after the stretching are presented in Fig. 7. Initially the gold film was continuous and smooth. Riston island imprint in PDMS is clearly visible. At 19% strain, the gold stripe on bare PDMS near the island edges clearly buckled because of the lateral contraction of the substrate [Fig. 7(b)]. The gold film on the island and in between the islands remained smooth. Around and at the island edges, the gold film delaminates from the substrate but electrical continuity remains. After release [Fig. 7(c)], the Au surface under the optical microscope is very similar to its initial state. A few cracks are visible at the bonded/delaminated gold transition. SEM observation revealed that the Au film between the islands contains a network of micron scale cracks of the type reported earlier.²⁵ The Au film on the DLC islands was smooth and conformed to the DLC topography. Cracks perpendicular to the stretching direction were indeed observed at the island edges, but did not propagate across the width of the gold stripe. Probably these cracks result from strain concentration at the island edge, as predicted by the FEM simulation.

VI. DISCUSSION

We have investigated two other island materials, SiO_x and SiN_x . Arrays of 400 nm thick SiO_x islands were prepared by electron-beam evaporation on PDMS substrate. Geometrical parameters were $a=b=100 \mu\text{m}$. Large wrinkles (wavelength $\sim 20 \mu\text{m}$, amplitude $\sim 1 \mu\text{m}$) were observed in the oxide film. The samples were stretched using a procedure similar to the one described above. Upon stretching, SiO_x islands first deformed elastically then cracked at $\epsilon_{\text{appl}} \sim 9\%$. We found that the strain in between the islands was about twice the applied strain, as predicted by Eq. (2). Though the island strain was greatly reduced compared to the applied strain, SiO_x islands on PDMS are not stiff enough and cannot prevent cracking of additional device materials upon large deformation.

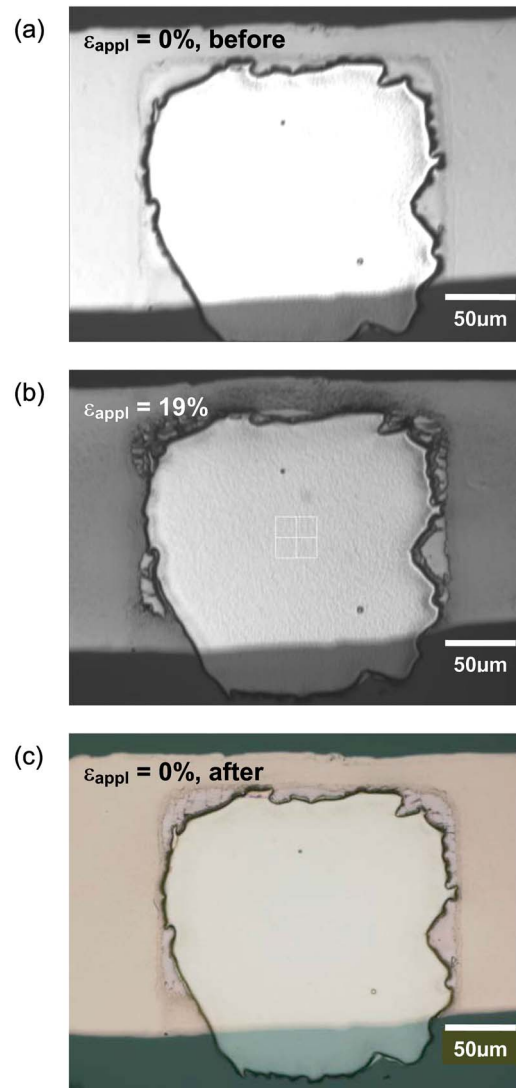


FIG. 7. (Color online) Optical micrograph of a gold interconnect running over one DLC island on PDMS substrates: (a) before deformation; (b) while stretched by 19% tensile strain; and (c) after release of the mechanical loading.

We have also studied PECVD grown SiN_x islands on PDMS. SiN_x islands were 2 mm diameter and spaced 4 mm apart. Most of the islands were wrinkled with a small wavelength of $< 2 \mu\text{m}$. A test run was performed on a 1 mm wide gold interconnect patterned over five SiN_x islands. The islands cracked quickly upon stretching to $\epsilon_{\text{appl}} \leq 1\%$. Despite long transversal cracks across the nitride islands, the gold conductor maintained electrical conduction up to $\epsilon_{\text{appl}} = 10\%$. SiN_x islands smaller than 1 mm diameter need further study in view of the importance of $\text{SiN}_x/\text{a-Si:H}$ (amorphous silicon) in large-area electronics.

Experiments described in this paper validate the architecture of stretchable electronics presented in Sec. II, based on a compliant elastomeric substrate, stiff subcircuit islands, and stretchable interconnects. Although the present design and materials need further experimentation, we can derive the following guidelines to prepare electronic systems robust to large and elastic mechanical deformation.

First, the substrate should be highly compliant, with a

typical Young modulus of the order of 1 MPa, to divert most of the applied elongation away from the subcircuit islands and ensure the system elasticity.

Second, the strain in the islands should be kept below 1%, the critical fracture strain of most device materials. To do so, the island material should be “hard,” almost nondeformable, and stiffer than the substrate. We have shown that among tested materials, DLC films formed the stiffest islands. Also the island size should not be larger than the substrate thickness.

Third, small island fill factor, i.e., $a/b < 1$, favor smaller interisland strain ensuring interconnect integrity.

Finally, stretchable interconnects running on the elastomeric substrate and over several subcircuit islands are more resistive than on glass or plastic substrates. However, their electrical resistance remains fully compatible with thin-film transistor impedance (in the megaohm range) as well as switching frequencies required for most sensor array applications envisioned for stretchable electronics ($f < 100$ kHz).

VII. CONCLUSION

We are now able to integrate stiff subcircuit islands on elastomeric substrate and can interconnect them with stretchable metallization. Among DLC, SiO_x , and SiN_x , diamondlike carbon islands provide the stiffest platforms. The results presented in this paper provide fundamental elements for fully integrated stretchable electronics.

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