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Two-Dimensional Stretchable Organic Light-Emitting Devices with High Efficiency

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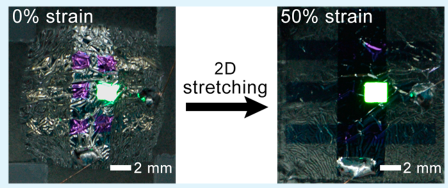
Supporting Information

ABSTRACT: Stretchable organic light-emitting devices (SOLEDs) with two-dimensional (2D) stretchability are superior to one-dimensional (1D) SOLEDs in most practical applications such as wearable electronics and electronic skins and therefore attract a great deal of interest. However, the luminous efficiency of the 2D SOLEDs is still not practical for the purposes of commercial applications. This is due to the limitations on materials and structures from the physical and electrical damage caused by the complicated interactions of the anisotropic stress in 2D stretchable system. Here 2D SOLEDs with excellent stretchability and electroluminescence performance have been demonstrated based on an ultrathin and ultraflexible OLED and a buckling process. The devices endure tensile strain of 50% in area with a maximum efficiency of 79 cd A^{-1}, which is the largest luminescent efficiency of 2D SOLEDs reported to date. The 2D SOLEDs survive continuous cyclic stretching and exhibit slight performance variations at different strain values. The 2D SOLEDs reported here have exhibited enormous potential for various practical applications.



KEYWORDS: *organic light-emitting devices, two-dimensional stretchable, ultraflexible, high-efficiency, mechanical robustness*

INTRODUCTION

Stretchable organic light-emitting devices (SOLEDs) have achieved lots of attentions in recent years and can be integrated with stretchable integrated circuits,[1] conductors,[2−4] transistors,[5−7] solar cells[8−10] and capacitors[11,12] to form functional and stretchable electronic systems. Compared with conventional light-emitting devices based on rigid or nonelastic substrates, SOLEDs could be conformable to complex surface topology and play a more and more important role in some emerging applications such as wearable electronics and electronic skins. Previous reports and demonstrations mainly focused on one-dimensional (1D) stretchable devices with large stretchability, high efficiency, and mechanical robustness.[13−17] SOLEDs with two-dimensional (2D) stretchability could greatly expand their practicability, especially for applications associated with human body and robotics which demand multidimensional stretchability. Some previous reports have exhibited stretchable devices with 2D stretchability based on different stretching strategies, but few researchers have made a detailed study on the electroluminescent (EL) performance of these devices under 2D stretching states. Discrete rigid light-emitting units have been combined with stretchable electrical interconnections to form a hybrid structure with 2D stretchability.[18−20] Stretchable light-emitting devices with intrinsic stretchability have been fabricated, in which the component layers and materials must be highly elastic.[14,15,21] Although the reported devices are well-designed and exhibit 2D stretchability, their original efficiency is much less than the required values for commercial applications and their EL performance under 2D stretching state is not investigated yet. The materials used and the structure design of the devices limit the original efficiency. The physical and electrical damage to the devices caused by the complicated interactions of the anisotropic stress under 2D stretching is more easily generated than 1D stretching and thus makes the EL performance of these stretchable light-emitting devices degrade quickly with 2D strain increasing.

Here we report a SOLED with high efficiency and excellent 2D stretchability. Random networks of buckles have been formed within the device by adhering an ultrathin and ultraflexible OLED onto a prestretched adhesive and elastomeric substrate. The 2D SOLEDs can bear tensile strain of 50% in area by biaxial stretching. The EL performance of the 2D SOLEDs has been investigated under different 2D stretching states for the first time among all the reported 2D SOLEDs. The 2D SOLEDs exhibit efficiency of 79 cd A^{-1} under 50% strain, which is the largest efficiency to date to the best of our knowledge. The 2D SOLEDs show stable EL performance at different strain values and can survive 100 times of 2D cyclic stretching. This is an important step toward commercially viable applications for the 2D SOLEDs in the near future.

EXPERIMENTAL SECTION

Ultrathin OLEDs Fabrication. Si substrates (2 cm × 2 cm) were subjected to ultrasonic cleaning with acetone, ethyl alcohol, and deionized water in sequence and then dried in a drying oven. Octadecyltrichlorosilane (OTS) modification was performed on the

Received: August 17, 2016
Accepted: October 28, 2016
Published: October 28, 2016

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DOI: 10.1021/acsami.6b10328
ACS Appl. Mater. Interfaces 2016, 8, 31166−31171

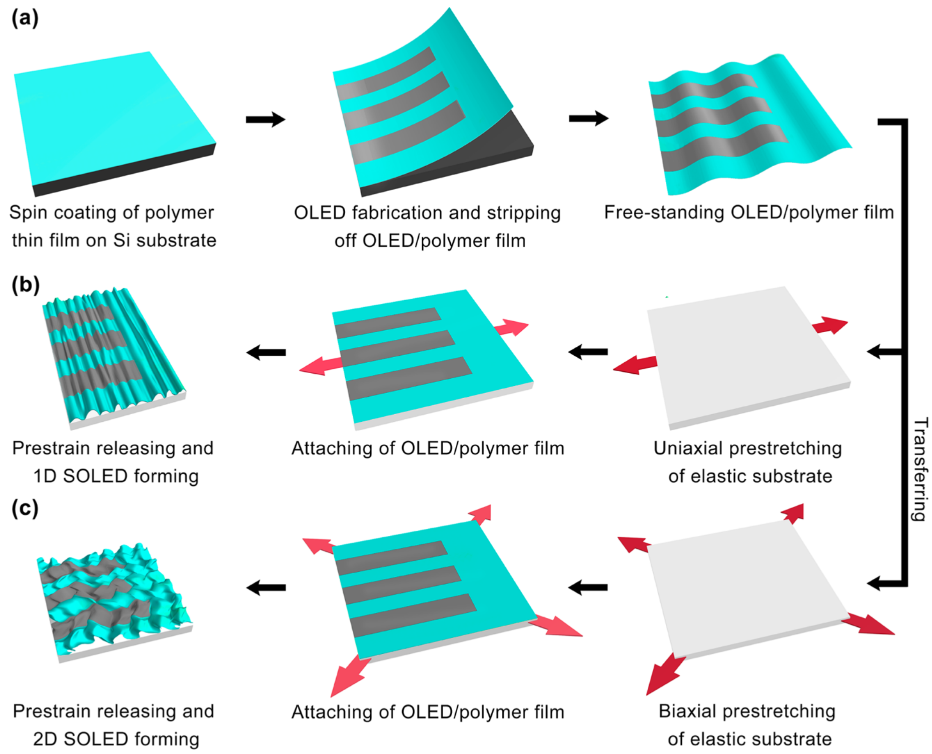

Figure 1. Schematic of the fabrication process of SOLEDs. (a) Fabrication of the ultrathin OLED: spin-coating photopolymer thin film on the Si substrate and curing by UV exposure, fabricating OLEDs on the polymer film, and stripping off the OLED/polymer film from the Si support to obtain a free-standing ultrathin OLED. (b) 1D SOLED fabrication: Transferring and adhering the ultrathin OLED on the uniaxially prestretched elastic substrate and 1D SOLEDs forming after releasing the prestrain. (c) 2D SOLED fabrication: Transferring and adhering the ultrathin OLED on the biaxially prestretched elastic substrate and 2D SOLEDs forming after releasing the prestrain.

pretreated Si substrates using the same method reported in our previous work.[22] An ultrathin layer of photopolymer (NOA63, Norland Products, Inc.) was spin-coated onto the OTS-treated surface of the Si substrate at 12 000 rpm for 90 s and then with ultraviolet curing. We fabricated small-molecule based OLEDs on the Si substrate-supported polymer film by thermal evaporation in a high-vacuum chamber. The device structure is Ag (80 nm) as anode, MoO_3 (3 nm) as anodic modification layer, *N*,*N*′-diphenyl-*N*,*N*′-bis (1,1′-biphenyl)-4,4′-diamine (NPB) (40 nm) as hole transporting layer, tris(2-phenylpyridine)iridium(III) ($Ir(ppy)_3$) doped *N*,*N*′-dicarbazol-yl-3,5-benzene (mCP) (6%, 20 nm) as emitting layer, 1,3,5-tris(*N*-phenyl-ben-zimidazol-2-yl)benzene (TPBi) (35 nm) as electron transporting layer, and Ca (3 nm)/Ag (18 nm) as cathode. Finally, an ultrathin and flexible OLED was achieved by peeling off the OLED/polymer film from the Si support.

Stretchable OLEDs Fabrication. Adhesive and elastic substrates (3 M VHB tapes) were stretched uniaxially to 200% strain for 1D SOLEDs and stretched biaxially to 100% strain at both diagonal directions for 2D SOLEDs by lab-made moving stages (Figure S1). The ultrathin OLED was then put and adhered onto the prestretched elastic substrate. Stretchable OLEDs formed random buckles after releasing the prestretched elastic substrate. For the 2D SOLEDs, the prestretched elastic substrate was released by switching back the rotary knobs of the moving stage in sequence. Eutectic gallium−indium (EGaIn) was selected to make electrical contact due to its excellent properties, such as liquid at room temperature,[23] moldable,[24] and low resistivity (29.4×10^{-6} Ω·cm).[25] Copper wires were used to connect the EGaIn drops and the power source.

Characterizations. All cyclic stretching processes were done on homemade moving stages. The SEM images of the stretchable devices were taken with a JEOL JSM-7500F scanning electron microscope (JEOL Ltd.). Dimension Icon AFM (Bruker Corporation) was used for taking the atom force microscopy images. XP-2 stylus profilometer (Ambios Technology, Inc.) was selected to measure the thickness of the ultrathin polymer film. The *I*−*L*−*V* characteristics of all OLEDs were investigated by a Keithley 2400 source meter and a Photo research PR-655 spectrophotometer. All measurements were carried out at room temperature in air without encapsulation.

RESULTS AND DISCUSSION

The schematic diagram of the fabrication procedure for the SOLEDs is shown in Figure 1. A template stripping technique was used to fabricate the free-standing ultrathin OLEDs. OTS modification was performed on a precleaned silicon (Si) substrate to realize a hydrophobic surface with lowered surface energy (Figure S2), which was beneficial to the template stripping process.[22,26] An ultrathin polymer film was obtained by spin-coating a photopolymer on the OTS-treated Si support and curing with ultraviolet (UV) exposure (Figure 1a, left). Its thickness is only 2.8 μm, which makes it very flexible. At the same time, the film is smooth with a root-mean-square (RMS) roughness of 0.36 nm (Figure S3). Small-molecule OLEDs were fabricated on the ultrathin polymer substrate by thermal evaporation in a high-vacuum chamber. Then, the ultrathin OLED was stripped off from the Si support entirely (Figure 1a, middle). The obtained free-standing ultrathin OLED was complete without any defects due to the lowered surface energy of the OTS-treated Si substrate (Figures 1a, right, and S4). Finally, the SOLED with random buckles was formed by transferring and adhering the ultrathin OLED onto the prestrained adhesive and elastic substrate and then releasing the prestrain on the elastic substrate. Both 1D (Figure 1b) and 2D (Figure 1c) SOLEDs can be obtained by prestretching the elastic substrates in uniaxial and biaxial directions, respectively.

It can be seen from Figure 1 that the stretchability of the SOLEDs is obtained by forming buckles. The ultrathin OLEDs

DOI: 10.1021/acsami.6b10328
ACS Appl. Mater. Interfaces 2016, 8, 31166−31171

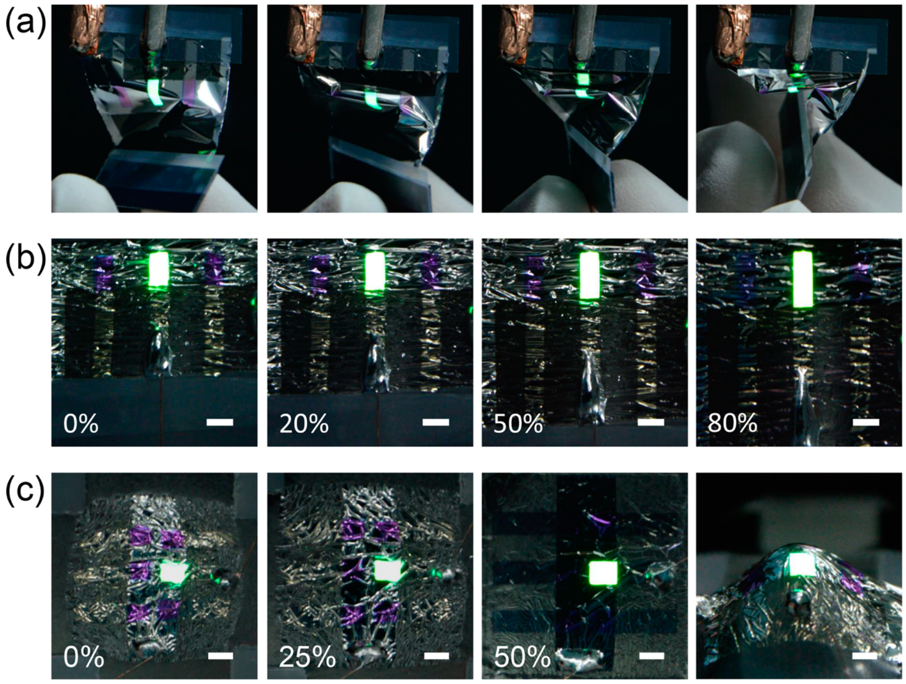

Figure 2. (a) Ultraflexibility of a free-standing ultrathin OLED demonstrated by buckling and twisting. The emitting area is 1.5 × 3.5 mm^2. The driving voltage is 4 V. (b) Images of a 1D SOLED operating at 5 V at strains of 0, 20, 50, and80%. Scale bar, 2 mm. (c) Images of a 2D SOLED operating at 5 V at strains of 0, 25, and50% in area and spreading over a 1 cm diameter centrifuge tube with three-dimensional deformation (right). Scale bar, 2 mm.

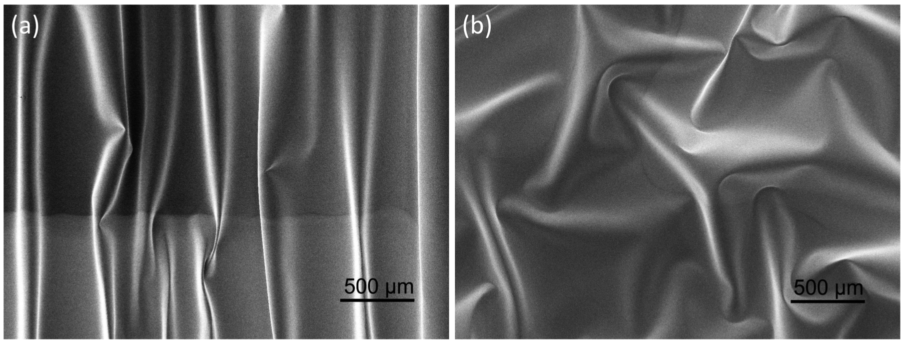

Figure 3. SEM images of the (a) 1D SOLEDs and (b) 2D SOLEDs at 0% tensile strain.

bent at the buckling regions during the stretch−release process, making the entire device stretchable. This basic concept of converting stretching to buckling was originally published for silicon-based electronics.[1] The high flexibility of the ultrathin OLEDs is important, because the buckles with small bending radii are required for applications such as wearable electronics and electronic skins. It is essential for the flexible OLEDs to survive extreme deformations such as twisting, folding, buckling, and crumpling without disastrous failure. To demonstrate that the ultrathin OLED is able to bear such shape changes, we suspended our device between two pieces of Si slice as seen in Figure 2a and Movie S1.

One of the Si slices was then moved closer to the other and twisted by 90° simultaneously, during which the device was bent, buckled, and twisted. While the ultrathin OLED worked continuously during several cycles of this deforming process, demonstrating its ultraflexibility. The EL performance of the ultrathin and ultraflexible OLEDs at planar state (with Si substrate as support) was measured. Small-molecule material of tris(2-phenylpyridine)iridium(III) (Ir(ppy)$_3$) was used as the emitter. The corresponding current density−luminance−voltage and current efficiency−voltage characteristics of the planar device are shown in Figure S5. The turn-on voltage is 3 V. The largest current efficiency is 71 cd A^{-1} at 4 V with a luminance value of 205 cd m^{-2}.

The stretchability of the 1D SOLEDs was first examined and is shown in Figure 2b. With 200% prestrain on the elastic substrate, the obtained 1D SOLEDs exhibited a maximum tensile strain of about 80%. A random network of buckles can be observed after releasing the prestrain. Uniform light emission can be observed across the entire luminous zone during the cyclic stretching test (Movie S2). Figure 3a shows the scanning electron microscopy (SEM) image of the 1D SOLEDs at the 0% tensile strain state (the largest compressive strain state). The bending radii of the buckles are estimated to be about 30−100 μm. The OLEDs on the top surface of the ultrathin polymer substrate are in tension at this bending state. The bending strain (S) can be simply expressed as

DOI: 10.1021/acsami.6b10328
ACS Appl. Mater. Interfaces 2016, 8, 31166−31171

$$S = \frac{t_L + t_S}{2R}\frac{1 + 2\eta + \chi\eta^2}{(1 + \eta)(1 + \chi\eta)} \quad (1)$$

where t_L is the thickness of the OLEDs, t_S is the thickness of the polymer substrate, $\eta = \frac{t_L}{t_S}$, R is the bending radius, $\chi = \frac{E_L}{E_S}$, E_L is the Young's modulus of OLED materials, and E_S is the Young's modulus of the ultrathin polymer substrate.[27] It can be noted that the bending strain is affected directly by the bending radius and the thickness of the polymer substrate. To achieve small bending radius, ultrathin polymer substrates are needed to avoid large bending strain. The stretchable OLED has a multilayered structure with the metallic anode contacting directly with the polymer substrate. The bending strain in the metallic anode is about 2% when the bending radius is 30 μm (t_L = 80 nm, E_L = 63 GPa; t_S = 2.8 μm, E_S = 1.6 GPa) where the influence of the organic layers is ignored due to the weak adhesive force between the metallic anode and the organic layers. The OLEDs do not crack or delaminate despite having a complex and multilayered structure and remain working efficiently at this strain value. This can be attributed to the strong support of the ultrathin and ultraflexible polymer substrate. The EL performance of the 1D SOLEDs has also been examined as shown in Figure S5. It can be seen that the 1D SOLEDs show comparable EL performance under various levels of tensile strain with that of the planar devices. The excellent and stable EL performance of the 1D SOLEDs indicates that the small bending radii of the buckles cause no mechanical or electrical damages to the ultrathin OLEDs. To investigate the durability of the devices under repeated stretching, we cycled the device from 0 to 40% strain with 1000 stretch−release cycles as shown in Figure S6. The mechanical stability of 1D SOLEDs during cyclic stretching measurements further demonstrates the ultraflexibility of the ultrathin OLEDs.

With such ultraflexible OLEDs, we successfully fabricated 2D SOLEDs by expanding the prestretching of the elastic substrate from uniaxial direction to biaxial directions. With 100% prestrain at both diagonal directions on the elastic substrate, we obtained 2D SOLEDs with 50% tensile strain in area as shown in Figure 2c. Uniform light emission from the entire luminous zone has been observed under repeated stretch−release cycles (Movie S3). Random buckles with small bending radii and a much more complicated topology were formed on the devices (Figure 3b). This is due to the interaction of the orthogonal compressive stress on the ultrathin OLED when releasing the prestrain. Stretchable electronics may offer unrestricted conformability with arbitrary curved surfaces, such as the hemispherical substrate of an electronic eye camera[28] and the end of a pencil,[15,19] which will require multidimensional deformability. The good conformability of our 2D SOLEDs was demonstrated by deforming the devices with a 1 cm diameter centrifuge tube (right in Figure 2c). The 2D stretchable device was functional when pressed from 0% strain state to about 50% strain state at multidirections and successfully spread over the top surface of the centrifuge tube.

Figure 4 shows the EL performance of the 2D SOLEDs. It can be seen that the 2D SOLEDs exhibit similar EL performance at different strain values with planar devices and 1D SOLEDs described above. The maximum luminance is 8080, 8137, and 9699 cd m^{-2} at 6.5 V (Figure 4a) and the maximum efficiency is 62.9, 72, and 79 cd A^{-1} at 3.5 V (Figure 4b) at strain of 0, 25, and 50% in area, respectively. The

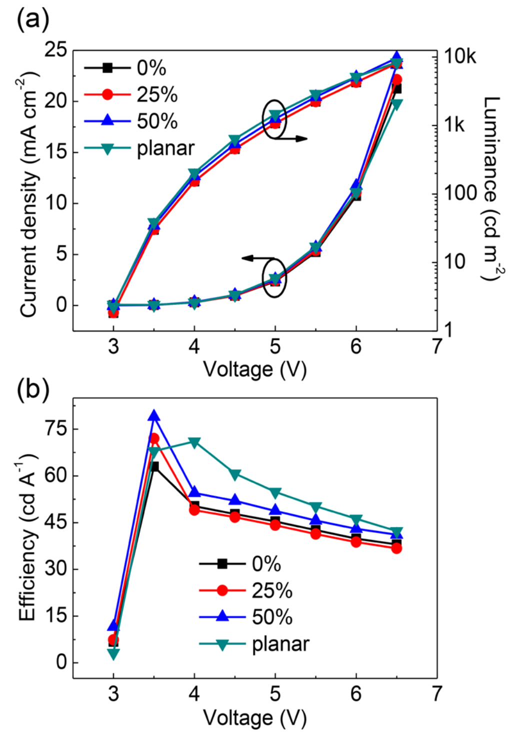

Figure 4. EL characteristics of the 2D SOLEDs. (a) Current density−luminance−voltage curves and (b) current efficiency−voltage curves of 2D SOLEDs with different strain values in area. A planar device is examined for comparison.

maximum efficiency of the devices increased with stretching. This may be attributed to the morphology changing of the emission area as strain increasing. The EL spectra are measured and shown in Figure S7. Narrowing and redshifting of the peak wavelength of the spectra with increased strain can be observed. This is due to the microcavity effect caused by the metallic bottom and top electrodes. The photon density of states within the microcavity is redistributed, which results in the angular dependence of the emission. Only certain wavelengths, which conform to the cavity modes, can be emitted in given directions. The relationship between the wavelength of the cavity modes and the emission direction can be written by

$$2nd\cos(\theta) = m\lambda \quad (m = 1, 2, ...) \quad (2)$$

where λ is the resonance wavelength, m is the mode number, n is the refractive index of the organic layers, d is the distance between the two metallic electrodes, and θ represents the emission angle which is related to the viewing angle through Snell's law.[29,30] Equation 2 shows that the resonance wavelength increases with the decrease of θ, which means that redshift of the EL spectra occurs with decreased viewing angle.

At 0% tensile strain, the ultrathin OLEDs have the smallest bending radii and the largest viewing angles at the buckling regions. Therefore, blueshift of the EL spectra can be observed. It can be seen from Figure 3b that the 2D buckles are heterogeneous which means that different bending radii and viewing angles exist at the same time. The amount of blueshift of the EL spectra at different emission zones is different, so the EL spectra are broadened compared with those of the planar devices. With the increasing of the tensile strain, the bending radii of the buckles increase, which results in the decrease of the viewing angles. Narrowing and redshift of the EL spectra can be observed. At the largest tensile strain, the ultrathin OLEDs are

DOI: 10.1021/acsami.6b10328
ACS Appl. Mater. Interfaces 2016, 8, 31166−31171

approximately flat and nearly have the same EL spectra with the planar device.

The mechanical stability of 2D SOLEDs is crucial for their applications and was examined under repeated stretch−release cycles. Figure 5 shows the characteristics of 2D SOLEDs under cyclic stretching. All measurements were conducted at 0% tensile strain during this testing process. There are only about 5.8 and 7.5% degradations for the current density and luminance after 100 stretch−release cycles with strains between 0 and 25%, respectively (Figure 5a). As a consequence, the current efficiency degraded 1.7% after the cyclic stretching test (Figure 5b). These results show that the 2D SOLEDs reported here are mechanically robust, which can be attributed to the ultraflexibility of the ultrathin OLEDs.

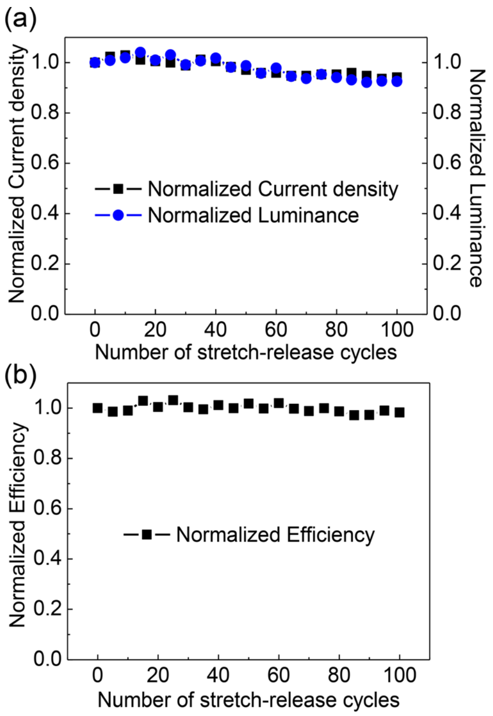

Figure 5. Mechanical robustness characterization of the 2D SOLEDs. (a) Normalized current density and luminance and (b) normalized current efficiency of the 2D SOLEDs under 100 stretch−release cycles between 0 and 25% strain in area. The bias voltage is 5 V, and all EL measurements were conducted at 0% strain in area.

CONCLUSIONS

We have fabricated 2D SOLEDs with excellent stretchability and EL performance by a simple buckling process. The ultrathin and ultrasmooth polymer substrate, fabricated simply by spin-coating process, provides the OLEDs with strong support and ultraflexibility. The flexibility for materials selected and structure design of the device guarantees the high efficiency of the SOLEDs. The resulting 2D SOLEDs can bear tensile strain of 50% in area by biaxial stretching and show uniform light emission and high current efficiency of >70 cd A^{-1} at different strain values. The 2D SOLEDs exhibit good conformability and successfully spread over a curved surface. The mechanical stability of the 2D SOLED has been examined by cyclic stretching during which the device only shows a small degradation in EL performance. This is the first report of a 2D SOLED with high EL performance. This simple buckling process is compatible with thermal evaporation of metals and organic materials and spin-coating, as well as lots of other processing technologies. This work makes a progress on the road of realizing flexible, stretchable, and surface-conforming electronic devices and may lead to stretchable OLEDs becoming commercially viable in the near future.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.6b10328.

Photographs of homemade moving stages for 1D and 2D stretching. Images of contact angle measurements of untreated Si substrate and OTS-treated Si substrate. AFM image of the ultrathin polymer film. Image of free-standing ultrathin OLEDs. Current density−luminance−voltage and efficiency−voltage characteristics of the 1D SOLEDs at different strain values. EL performance of the 1D SOLEDs under 1000 stretch−release cycles between 0 and 40% strain. EL spectra of the 2D SOLEDs at different stretching states. (PDF)

Movie of exhibition of free-standing ultrathin OLEDs (AVI)

Movie of exhibition of ultrathin 1D SOLEDs (AVI)

Movie of exhibition of ultrathin 2D SOLEDs (AVI)

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the National Nature Science Foundation of China (NSFC) and National Basic Research Program of China (973 Program) under Grant Nos. 61322402, 61590930, 61505065, 61435005, and 2013CBA01700.

ABBREVIATIONS

SOLEDs, stretchable organic light-emitting devices
2D, two-dimensional
1D, one-dimensional
EL, electroluminescent
OTS, octadecyltrichlorosilane
RMS, root-mean-square

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DOI: 10.1021/acsami.6b10328
ACS Appl. Mater. Interfaces 2016, 8, 31166−31171