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## <sup>1</sup> Standing Enokitake-like Nanowire Films for <sup>2</sup>Highly Stretchable Elastronics

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#### 9 **S** [Supporting Information](#page-5-0)

 ABSTRACT: Stretchable electronics may enable electronic 11 components to be part of our organs-ideal for future wearable/implantable biodiagnostic systems. One of key challenges is failure of the soft/rigid material interface due to mismatching Young's moduli, which limits stretchability 15 and durability of current systems. Here, we show that standing enokitake-like gold-nanowire-based films chemi-17 cally bonded to an elastomer can be stretched up to 900% and are highly durable, with >93% conductivity recovery even after 2000 stretching/releasing cycles to 800% strain.



<sup>20</sup> Both experimental and modeling reveal that this superior elastic property originates from standing enokitake-like <sup>21</sup> nanowire film structures. The closely packed nanoparticle layer sticks to the top of the nanowires, which easily cracks 22 under strain, whereas the bottom part of the nanowires is compliant with substrate deformation. This leads to tiny V-

<sup>23</sup> shaped cracks with a maintained electron transport pathway rather than large U-shaped cracks that are frequently

<sup>24</sup> observed for conventional metal films. We further show that our standing nanowire films can serve as current collectors in

<sup>25</sup> supercapacitors and second skin-like smart masks for facial expression detection.

<sup>26</sup> KEYWORDS: standing nanowire film, unconventional crack, elastronics, electronic skins, strain sensors

27 electronics are transitioning from the current rigid<br>29 version to a next-generation flexible design, which will<br>30 elastronics). In an elastronic system, its components can be version to a next-generation flexible design, which will **ultimately** evolve into stretchable electronics (*i.e.*, seamlessly integrated with skin/muscles to become parts of our organs, thereby enabling genuine biodiagnostics in real time and in situ. It is well-known that elastronics requires a seamless combination of stretchability and electrical conductivity, which 35 can be achieved extrinsically or intrinsically.<sup>1–3</sup> The former is 36 achieved by designing structures that stretch,<sup>4-10</sup> whereas the latter is realized by producing materials th[at](#page-6-0) are deform-38 able.<sup>11−22</sup>

 An i[dea](#page-6-0)l elastronic system may be made from intrinsically elastic components, including conductors, resistors, diodes, transistors, and sensors, so that they can integrate with 42 modulus-matching skin/muscle,<sup>12-19,23,24</sup> ideal for wearable/ implantable diagnostics wit[h](#page-6-0) t[rue](#page-6-0) [ca](#page-6-0)pability of health monitoring anytime and anywhere. A viable strategy is to deposit active nanomaterials onto or embed them into elastomers.[12,14](#page-6-0)<sup>−</sup>17,25−<sup>33</sup> Among them, one-dimensional nano- materials are p[articu](#page-6-0)l[arl](#page-7-0)y promising as they can be used to construct percolation networks onto or into elastomeric

matrices.  $12,14,22,25-32,34$  Two-dimensional (2D) percolation 49 nanowir[e-based](#page-6-0) [th](#page-6-0)i[n](#page-6-0) [fi](#page-7-0)lms have demonstrated a wide range of <sup>50</sup> applications in wearable electronic skin (e-skin) sensors,  $35$  soft 51 energy devices,  $36,37$  organic light-emitting diodes,  $38$  m[emory](#page-7-0) 52 devices,  $39$  PM [2.5](#page-7-0) filters,  $40$  soft robotics,  $26$  and [transparent](#page-7-0) 53 electronics.  $41-46$  [Despite this encouraging](#page-7-0) progress, delamina-  $54$ tion and/[or](#page-7-0) c[rac](#page-7-0)ks at the soft/rigid materials' interface often <sup>55</sup> occur under large or repeated strains due to mismatching <sup>56</sup> Young's moduli between active rigid materials and soft <sup>57</sup> elastomeric matrixes. This limits the stretchability and long- <sup>58</sup> term durability of current systems, preventing them from being <sup>59</sup> used in real-world applications. $4\overline{6}$ 

In this work, we show tha[t](#page-7-0) standing enokitake-like gold <sup>61</sup> nanowire films chemically bonded to elastomeric materials can <sup>62</sup> exhibit stretchability (up to 900%) much higher than that of <sup>63</sup> conventional vacuum-evaporated bulk metal or percolating <sup>64</sup> nanowire films, without any additional extrinsic buckling <sup>65</sup> design. This was achieved because of standing enokitake-like 66

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Figure 1. Characteristics of standing enokitake-like nanowire-based gold films. (a−c) Optical appearances of standing enokitake-like nanowire-based films grown on flat elastomers: (a) PET, (b) PDMS, (c) Ecoflex. (d,e) Photographs of the thin standing enokitake-like nanowire-based gold films with skin-textured Ecoflex substrate on a human thumb knuckle while bending and releasing, respectively. (f,g) Typical top-view and side-view SEM image of standing enokitake-like nanowire-based gold films. Scale bar: 200 nm. (h−k) SEM images of standing nanowire films with different thicknesses: (h) ~1.5 μm, (i) ~3.5 μm, (j) ~7 μm, and (k) ~14 μm. Scale bar: 1 μm. (l) Change of nanowire height as a function of growth time.

 nanowire structures and their strong adhesion with elastomers, leading to distinct stretching behaviors. Unlike conventional metal films (by vacuum evaporation/sputtering or previous nanomaterials films) which typically exhibit large "cliff-like" "U-shaped" cracks that cannot recover upon releasing the strain, our standing enokitake-like nanowire gold films instead show tiny "V-shaped" cracks that are able to recover the conductivity when strain is removed. The formation of V- shaped cracks is due to hierarchical structures of the nanowire film, in which the top nanoparticle layer is mechanically more rigid than the underlying nanowire layer. This leads to initial cracking that starts from the top particle layer under low level of strains (typically below 300%), followed by conventional large U-shaped cracks of the entire film under large strains (typically between 300 and 800%). In both cases, conductivity pathways could be maintained. This unconventional property enables our enokitake-like nanowire film to be used as highly durable conductors which could retain the >93% conductance even after 2000 stretching/releasing cycles to 800% strain. We demonstrate specifically here that they can be applied to fabricate intrinsically stretchable supercapacitors and can be used as "second-skin" facial expression recognition mask <sup>89</sup> sensors.

#### 90 RESULTS AND DISCUSSION

 By extending the method of seed-mediated electroless plating 92 on rigid surfaces,<sup>48</sup> standing enokitake-like nanowire-based gold films could [grow on a number of polymer substrates](#page-7-0) including polyethylene terephthalate (PET), polydimethylsi- loxane (PDMS), and Ecoflex (highly stretchy silicone rubber). Macroscopically, the standing nanowire films were uniform with a shiny gold reflective surface if the underlying f1 98 elastomeric substrates were flat (Figure 1a−c). The fabrication process is illustrated in Figure S1. In brief, an elastomeric 100 substrate is first treated using  $O_2$  plasma to render its surface hydrophilic, which is then followed by silanization with (3- aminopropyl)trimethoxysilane (APTMS). Next, negatively charged seed particles could be immobilized onto this amine-functionalized surfaces via electrostatic attraction. Further

immersion of the seed-particle-modified elastomer into a <sup>105</sup> growth solution containing gold precursors, surfactants, and <sup>106</sup> reducing agents could lead to the formation of densely packed <sup>107</sup> standing nanowire arrays. The gold films grown on thin Ecoflex <sup>108</sup> sheets ( $\sim$ 20 µm thickness) could naturally attach to human 109 skin wrinkles before and after stretching (Movie S1). The <sup>110</sup> growth process was found to be scala[ble and a](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_001.avi)ble to <sup>111</sup> conformably coat a range of other polymer substrates from <sup>112</sup> macroscopic to microscopic (Figure S2a−f) and even to <sup>113</sup> textured skin replicas (Figure 1[d,e and](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) Figure S2g). Superior <sup>114</sup> skin conformal attachment in conju[nction with](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) chemical <sup>115</sup> inertness and biocompatibility of gold indicates the great <sup>116</sup> potential of our nanowire film as second skin patches for <sup>117</sup> various biomedical applications. 118

Further top-view and side-view characterizations by scanning <sup>119</sup> electron microscopy (SEM) revealed enokitake-like nanowire <sup>120</sup> film structures (Figure 1f,g), in which the top layer ("head"  $121$ side) consists of closely packed gold nanoparticles with a <sup>122</sup> diameter of 9.3  $\pm$  2.1 nm. The bottom layer ("tail" side) is 123 composed of nanowires standing normal to the elastomer <sup>124</sup> substrates, with a typical nanowire diameter of  $7.8 \pm 1.7$  nm. In 125 addition, the number density of nanowires can reach as high as <sup>126</sup> ~1.09  $\times$  10<sup>4</sup>  $\mu$ m<sup>-2</sup>, which is much higher than that of 127 previously reported 2D nanowire percolation network <sup>128</sup> systems.<sup>35–39</sup> The estimated porosity of the head side is 65− 129 72%, w[hereas](#page-7-0) the tail side is 50−55%. Longer growth times <sup>130</sup> lead to longer nanowires but reach the plateau in about 20 min <sup>131</sup> (SEM images in Figure 1h−k). We obtained nanowires that <sup>132</sup> were much longer than those in the literature<sup>48</sup> by using 133 concentrated growth solution to achieve tunable l[engths up to](#page-7-0) <sup>134</sup>  $∼15$  µm (Figure 11). In addition, the diameter of both 135 nanoparticle and nanowire did not change much as the <sup>136</sup> nanowire became longer (Figure S3). It is even possible to <sup>137</sup> grow staircase-like nanowire fi[lms by m](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)ask-assisted step growth <sup>138</sup> (Figure S4). Overall, the structural features including accurate <sup>139</sup> [height con](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)trol, standing enokitake-like configuration, and <sup>140</sup> control over surface topological structures indicate that our <sup>141</sup> system is different from a dominant nanowire percolation <sup>142</sup>



Figure 2. Superior intrinsic stretchability of standing enokitake-like nanowire-based gold films. (a) Comparison of stretchability among evaporated Au films, lying-down gold nanowire film and standing enokitake-like nanowire-based gold film. (b) Plot of normalized resistance  $(R/R_0)$  versus normalized length  $(L/L_0)$ . Scattered black squares denote experimental data; the red curve is the theoretical prediction based on the equation  $R/R_0 = (L/L_0)^2$ . Inset: Representative optical images of standing nanowire film under different strains of 0, 100, 250, and 300%. Scale bar: 20  $\mu$ m. (c) Comparison of this work to recent work in elastic conductors. Data points are extracted from the following papers: blue open triangle, Au nanoparticles (Au NPs);<sup>50</sup> pink open circle, Ag nanowires (Ag NWs);<sup>54</sup> black open square, Ag NWs;<sup>25</sup> lime open diamond, carbon nanotube  $(CNT)$ ;<sup>12</sup> black open pantagon, *in situ* Ag NPs;<sup>17</sup> [orange open pentagon, Ag nanoparticles \(Ag](#page-7-0) NPs);<sup>5</sup> pistachio open inverted triangle, Au nanosheets (Au NSs);<sup>40</sup> [Royal cross, carbon](#page-6-0) nanofibers (CNFs);<sup>57</sup> [green open pantagon, Ag](#page-6-0) flakes;<sup>[41](#page-7-0)</sup> sienna left open triangle, CNT;<sup>15</sup> purple open right triangle, Ag carbon nanotubes (Ag CNT);<sup>40</sup> cyan o[pen circle, CNT;](#page-7-0)<sup>56</sup> red filled star, t[his](#page-7-0) study. (d) Conductance chang[e of standing enokitake-like nanowire-based](#page-6-0) film during 2000 [stretching/releasing cycles up to 800% strain.](#page-7-0)

143 network<sup>[26](#page-6-0)[,35](#page-7-0)–39,46</sup> and may be viewed as a three-dimensional <sup>144</sup> percolation s[ystem](#page-7-0).

 We systematically investigate stretchability of the standing nanowire-based film. When directly grown on Ecoflex 147 substrates with the nanowire chemically bound to surfaces, the films exhibit exceptionally high stretchability up to 800% of f2 149 strain (Figure 2a, red solid line). With additional Ecoflex encapsulation, the conductivity was observed to survive even at the 900% strain, which is almost the physical limit of the Ecoflex elastomer (Figure S5). The improved stretchability with Ecoflex enca[psulation m](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)ay be due to the enhanced bonding at the top side, leading to more uniform crack propagation of the nanoparticle, preventing catastrophic failure. This observation is in agreement with sandwiched 157 silver-nanowire-percolated structure reported previously.<sup>49</sup> Remarkably, the original conductivity could be recover[ed](#page-7-0) upon stress release (Figure 2a, red dashed line). In control experiments, we found that the evaporated gold can only survive ∼10% strain before conductivity is lost, and the percolation lying-down nanowire film is only able to tolerate a ∼150% strain (blue solid line in Figure 2a). Both bulk metal and percolation nanowire films show no conductivity recovery upon stress release (black and blue dashed lines in Figure 2a). 166 We further plot normalized resistance  $(R/R_0)$  v*ersus* 167 normalized length square  $(L/L_0)^2$  for experimental data 168 collection and theoretical prediction (Figure 2b), where  $R_0$  and  $L_0$  are the resistance and length, respectively, of samples at 0% strain. The deviation starts at a strain of ∼150%, above

which cracks form and propagate, which is further validated <sup>171</sup> from optical imaging (inset of Figure 2b). This threshold value <sup>172</sup> is 3-fold that for copper-bonded Kapton film.<sup> $4/$ </sup> Note that  $173$ 800% stretchability for an enokitake-like stand[ing nanowire](#page-7-0) <sup>174</sup> film outperforms the state-of-the-art inorganic stretchable <sup>175</sup> conducting film  $12,15,17,25,50-57$  (Figure 2c). Remarkably, the 176 film conductanc[e](#page-6-0) [G](#page-6-0) [retai](#page-6-0)[ned](#page-7-0)  $>93\%$  $>93\%$  of the initial conductance 177  $(G<sub>0</sub>)$  after stretching/releasing to 800% strain for 2000 cycles 178 (Figure 2d). This has not yet been achieved, to the best of our <sup>179</sup> knowledge, by previously reported stretchable conductors <sup>180</sup> without using prestrain or buckling designs. 181

We further established that strong adhesion between the <sup>182</sup> nanowire and Ecoflex substrate and "accordion-fan-like" V- <sup>183</sup> shaped cracking processes is responsible for the exceptional <sup>184</sup> high stretchability observed. The adhesion test (Movie S2) 185 clearly shows that our standing enokitake-like na[nowire](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_002.avi) film <sup>186</sup> could survive in the normal Scotch tape test multiple times <sup>187</sup> without significant resistance change. The strong adhesion may <sup>188</sup> be due to the use of APTMS that serves a bifunctional <sup>189</sup> molecular glue. Its amine side strongly interacts with gold <sup>190</sup> nanowires, and its silane sides covalently bond to Ecoflex <sup>191</sup> surfaces. The introduction of an organic intermediate layer has <sup>192</sup> been demonstrated as an effective strategy to improve the <sup>193</sup> adhesion between the metallic layer and polymeric substrates, <sup>194</sup> thus enhancing the overall performance of the stretchable <sup>195</sup> conductive film.  $58-61$  Unlike the continuous bulk metal film, 196 our nanophased [enok](#page-7-0)itake-like structures offer better stretch- <sup>197</sup> ability [\(Table](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) [S1\)](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf). 198



Figure 3. Optical microscopic and AFM characteristics of three different gold films (evaporated gold film, lying-down nanowire gold film, and standing enokitake-like nanowire-based gold film). Microscopic behavior of (a) evaporated Au film, (c) lying-down nanowire gold film, and (e) standing enokitake-like nanowire-based gold film by optical microscope imaging at various strain (from 0, 300, and back to 0%), respectively. AFM images and height plots of (b) evaporated gold film, (d) lying-down nanowire gold film, and (f) standing enokitake-like nanowire-based gold film under 300% strain. Nanowire height for standing enokitake-like nanowire-based gold film is 1.5 μm. Scale bar: 200  $\mu$ m. All optical images have the same resolution.

 Unlike conventional bulk gold or percolation nanowire films, our standing nanowire films have hierarchical structures with close-packed nanoparticle arrays on the top and aligned nanowires chemically bound to elastomeric substrates. This leads to a distinct stretching mechanism (Figure S6). For further investigation, we carried out de[tailed mul](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)tiscale morphological studies in order to understand the exceptional stretchability observed. We scrutinized morphological features in different locations of rectangle standing nanowire metallic nanopatches under various strains by optical microscopy (Figure S7). This offers a panoramic overview of our standing [nanowire](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) film stretching process at millimeter and micrometer length scales. Evident cracks will not be seen until about 300% strain is applied. At the nanoscale, atomic force microscopy (AFM) and cross-sectional SEM characterization under a stretched state clearly show the presence of V-shaped cracks (Figures S8 and S9). The cracking depths measured for the t[wo particular stand](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)ing nanowire films under different strains were significantly lower than the film thickness. Assuming that the nanowire deforms elastically without breaking up and with its ends firmly attached to elastomeric substrates, we can visualize a V-shaped cracking process by finite element analysis (Movie  $S3$ ). However, both bulk gold films and percolation f3 222 [nanowire](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_003.avi) films exhibit only typical U-shaped cracks (Figure f3 223 3a−d; also see Figure S10 for the schematic illustration of V- shaped crack a[nd U-shaped](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) crack). Both can tolerate a level of strain much less than that for the standing nanowire films. The concurrent film delamination prevents recovery of original structures, hence, leading to poor conductivity recovery (Figure S6a,b). Note that the stretching mechanism of our [nanowire](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) film is fundamentally different from previous aligned carbon nanotube arrays where building blocks were not

standing normal to the substrate but were lying down flush on <sup>231</sup> the substrate. $^{30}$  3232

The above [m](#page-6-0)ultiscale structural characterizations and finite 233 elemental analysis reveal the following mechanistic insights. <sup>234</sup> Cracks initiate from the head side, which serve as unzipping <sup>235</sup> points for strongly bundling nanowire arrays, yet the <sup>236</sup> interacting nanowire tail ends deform conformably to the <sup>237</sup> substrate without cracking (Figure 3e,f and Figure S6c). At the 238 point when substrate elongation commenc[es, the mech](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)anically <sup>239</sup> rigid top gold nanoparticle layer (head side) cracks, which <sup>240</sup> triggers the formation of V-shaped cracks as the strain level is <sup>241</sup> increased by unzipping them from the top side. This typically <sup>242</sup> occurs when the strain level is less than ∼150% strain, where <sup>243</sup> no delamination occurs between substrates and our gold film at <sup>244</sup> this stage. Obvious wrinkles are observed in the middle part of <sup>245</sup> the standing nanowire film because of the Poisson ratio of <sup>246</sup> Ecoflex substrate (Figure S7, middle left). As the strain <sup>247</sup> increases further to [a certain th](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)reshold, large U-shaped cracks <sup>248</sup> form as a result of the standing nanowire film sliding/ <sup>249</sup> delaminating from the supporting elastomeric substrates. The <sup>250</sup> U-shaped cracks propagate as the strain level is further <sup>251</sup> increased; however, percolation conductive pathways are still <sup>252</sup> maintained until reaching a catastrophic failing point. The V- <sup>253</sup> shaped and U-shaped cracks coexist at the high strain levels <sup>254</sup> typically from 300 to 800%. The self-repairable cracks were <sup>255</sup> also demonstrated from more detailed SEM characterization. <sup>256</sup> By inspecting the same spot in a particular sample, negligible 257 morphological changes were observed before and after 60 000 <sup>258</sup> cycles of stretching/releasing to 185% strain (Figure S11). Its <sup>259</sup> excellent stretchability was maintained even a[fter 40 week](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)s of <sup>260</sup> storage in ambient conditions without encapsulation [\(Figure](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) <sup>261</sup>  $S12$ ). 262



Figure 4. Real-time facial expressions monitoring. (a) Schematic illustration of the detection system setup. (b) Schematic of standing enokitake-like nanowire-based gold film smart mask design according to nine facial muscle group movements caused by various emotions. (c) Mobile device interface for result reading. (d−h) Real-time monitoring of five different facial expressions of happy, sad, angry, surprise, and fear.

 We also found that the stretchability of the standing nanowire film showed a decreasing trend, whereas nanowire length increased (Figure S13a). As the nanowire length increased to 14  $\mu$ m, the fi[lm los](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)t conductivity at 80% strain, which is 10 times lower than that of the 1.5  $\mu$ m film. As expected, the overall nanowire/Ecoflex sheet became stiffer as the nanowire length increased (Figure S13b). This could be due to strong wire-to-wire inte[ractions amo](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)ng longer nano- wires, rendering nanowire films more rigid, approaching bulk gold mechanical properties.

 The facile growth of a standing nanowire film in conjunction with their outstanding performances indicates their suitability for soft electronics applications. As the first proof of concept, we demonstrate their use in soft, stretchable supercapacitors using our gold film with short nanowires. In a typical symmetrical layout, we were able to achieve excellent capacitive behavior (Figure S14), which also shows negligible changes over a wide [range of ap](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)plied tensile strain from 0 to 250%. The slight capacitance increase from 0 to 100% strain may be due to increased surface area of the nanowire unzipping process under strain. Further stretching beyond the 100% strain caused a very small decrease in the capacitance, retaining 84% of the original capacitance at a strain up to 250% (Figure S15a,b). This slight degradation of capacitance is poss[ibly due to th](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)e conductivity decreases of standing nanowire film electrodes and/or deformation of the electrolyte layers over stretching. Nevertheless, specific capacitance could be maintained by 99% after 200 stretch/ release cycles at the strain of 200%, suitable for wearable on-body energy storage devices ([Figure](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) [S15c,d](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)).

The excellent skin conformability of our standing nanowire <sup>293</sup> film enabled its use as e-skin smart nanopatches for detecting <sup>294</sup> childhood autism disorder. Note that the smart nanopatches <sup>295</sup> were fabricated by a strain-sensitive film from longer standing <sup>296</sup> nanowires. Instead of an optical approach used by the NODA <sup>297</sup> diagnostic tool available on Apple store, we used nine e-skin <sup>298</sup> nanopatches to monitor particular pieces of muscle/skin <sup>299</sup> stretching related to facial expression (Figure 4). Based on 300 f4 the information from the Facial Action Coding System <sup>301</sup> (FACS) library from Ekman's group, $62$  we could relate 302 electrical signals to the five different e[motional expressions](#page-7-0) <sup>303</sup> (happy, sad, angry, surprise, and fear) in a wireless manner <sup>304</sup> (Movie S4). Different facial expressions can be read from a <sup>305</sup> [mobile scre](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_004.avi)en in real time. 306

#### CONCLUSIONS <sup>307</sup>

In summary, we report the exceptional high stretchability and <sup>308</sup> durability of standing enokitake-like nanowire-based gold films, <sup>309</sup> which are unexpected in the context of current dominant <sup>310</sup> nanowire percolation network-based stretchable conductors. <sup>311</sup> Our results clearly reveal that this is attributed to standing <sup>312</sup> enokitake-like nanowire structures, vertically aligned config- <sup>313</sup> uration, and strong chemical bonding interactions between <sup>314</sup> standing nanowire films and elastomeric substrates. Together, <sup>315</sup> this leads to distinct elastic properties that have never been <sup>316</sup> observed for conventional bulk metal films or other nanoma- <sup>317</sup> terial networks (both vertically aligned and lying-down aligned <sup>318</sup> carbon-nanotube-based systems; see Table S2 in the <sup>319</sup> Supporting Information). We further [demonstr](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf)ate the <sup>320</sup> applications of our standing nanowire film in stretchable <sup>321</sup> supercapacitors and wearable e-skin sensors, beyond which we <sup>322</sup>

<span id="page-5-0"></span><sup>323</sup> may find a myriad of additional applications in future <sup>324</sup> elastronics.

#### 325 **METHODS**

**Chemicals.** Gold(III) chloride trihydrate ( $HAuCl<sub>4</sub>·3H<sub>2</sub>O$ , 99.9%), triisopropylsilane (99%), 4-mercaptobenzoic acid (MBA, 90%), APTMS, sodium citrate tribasic dihydrate (99.0%), L-ascorbic acid, 329 poly(vinyl alcohol) (PVA) powder,  $H_3PO_4$ , and ethanol (analytical grade) were purchased from Sigma-Aldrich. All solutions were prepared using deionized water (resistivity >18 MΩ·cm<sup>−</sup><sup>1</sup> ). All chemicals were used as received unless otherwise indicated. Conductive wires were purchased from Adafruit.

 Elastomeric Substrates. PDMS substrates were made by mixing Sylgard 184 silicone elastomer base and curing agent at a weight ratio of 10:1. The mixture was poured on a 6 in. flat-plate Petri dish using 0.5 mm height shims as spacers and cured at 65 °C for 2 h in an oven. Ecoflex substrates were made by pouring Ecoflex curable silicone fluid (Smooth-On Ecoflex 00-30) into a 6 in. flat-plate Petri dish and curing under room temperature for 4 h.

 Synthesis of Standing Gold Nanowire Films. A modified seed-342 mediated approach was used, as described in the literature.<sup>8</sup> First, 2 nm seed gold nanoparticles were synthesized. Briefly, 0.147 [mL of 34](#page-6-0) 344 mM sodium citrate was added into a conical flask with 20 mL of  $H_2O$ 345 under vigorous stirring. After 1 min, 600  $\mu$ L of ice-cold, freshly prepared 0.1 M NaBH4 solution was added with stirring. The solution turned brown immediately. The solution was then stirred for 5 min and stored at 4 °C until needed.

 To grow standing nanowires on substrates (e.g., Si wafer, Ecoflex),  $O_2$  plasma was applied to render the surfaces hydrophilic. Depending on the types of substrates, the plasma treatment time varied from 2 to 17 min. Then the substrates were functionalized with an amino group by silanization reaction with 5 mM APTMS solution for 1 h. APTMS- modified substrates were immersed into excess citrate-stabilized Au seed (3−5 nm) solution for 2 h to ensure the saturated adsorption of gold seeds, followed by rinsing with water four times to remove the weakly bound seed particles. Finally, seed-particle-anchored substrates 358 were in contact with a growth solution containing 980  $\mu$ M MBA, 12 mM HAuCl4, and 29 mM L-ascorbic acid, leading to the formation of standing nanowire films. The length of nanowires depended on the growth reaction time. Typical nanowire heights of ∼1.5, ∼3.5, ∼5, ∼7, 362 and ∼14  $\mu$ m were obtained by adjusting the growth time to 2, 4, 5, 8, and 15 min, respectively.

364 Lying-Down Gold Nanowire Films. HAuCl<sub>4</sub>.3H<sub>2</sub>O (44 mg) was added into 40 mL of hexane, followed by addition of 1.5 mL of oleylamine. After the gold salts were completely dissolved, 2.1 mL of triisopropylsilane was added into the above solution. The resulting solution was left to stand for 2 days without stirring at room temperature until the color turned from yellow to dark, indicating the formation of gold nanowires. The chemical residues were removed by repeated centrifugation and thorough washing using ethanol/hexane  $(3/1, v/v)$  and finally concentrated to a 2 mL stock solution in hexane. The lying-down gold nanowire films could then be obtained by a simple drop-casting approach.

<sup>375</sup> Vacuum-Evaporated Gold Film. A 100 nm gold film could be 376 obtained using an e-beam evaporator (Intlvac Nanochrome II, 10 377 kV).

378 Characterization. SEM imaging was carried out using a FEI Helios Nanolab 600 FIB-SEM operating at a voltage of 5 kV. The sheet resistances of the standing enokitake-like nanowire-based gold films were carried out on a Jandel four-point conductivity probe by using a linear arrayed four-point head. To test the electromechanical responses for strain and bending sensing, the two ends of the samples were attached to motorized moving stages (THORLABS model LTS150/M). Uniform stretching/bending cycles were applied by a computer-based user interface (Thorlabs APT user), and the current changes were measured by the Parstat 2273 electrochemical system (Princeton Applied Research). For the analysis of detailed point load or pressure responses, a computer-based user interface and a force sensor (ATI Nano17 force/torque sensor) and a Maxon Brushless

DC motor using a high-resolution quadrature encoder (15  $\mu$ m of 391 linear resolution) were used to apply an external point load or 392 pressure. Ecoflex with a thickness of 500  $\mu$ m was chosen as the 393 substrate of the standing nanowire film in a strain test. PET with a <sup>394</sup> thickness of 125  $\mu$ m was chosen as the substrate of the standing 395 nanowire film in a strain test. PDMS with a thickness of 1 mm was <sup>396</sup> chosen as the substrate of the standing nanowire film in a point load/ <sup>397</sup> pressure test. The reflectance (R) data were collected from a <sup>398</sup> PerkinElmer UV−vis−NIR spectrophotometer (Lambda 1050) with 399 an integrating sphere setup. 400

Simulation. The finite element analysis model was implemented 401 in the ABAQUS 6.14/Standard software. Ecoflex substrate was <sup>402</sup> meshed using structured hex elements, whereas gold nanowires were 403 used a tetrahedral elements. There were a total of 2640 linear 404 hexahedral elements in the Ecoflex substrate and 106 200 quadratic <sup>405</sup> tetrahedral elements in the gold nanowire section. The aspect ratio of 406 the gold nanowire was modeled at 100, with a length of 800 nm and a 407 diameter of 8 nm. The elastic modulus and Poisson's ratio are 400 408 kPa and 0.49 for the Ecoflex substrate and 70 GPa and 0.42 for <sup>409</sup> nanowire, respectively. The boundary conditions were set by fixing <sup>410</sup> the left end of Ecoflex substrate and stretching uniaxially to 800% <sup>411</sup> elongation. The contact condition between the nanowire layer and 412 Ecoflex substrate was assumed to be pinned using a tie constraint. <sup>413</sup>

Elastic Supercapacitors. The standing enokitake-like nanowire- 414 based gold film was cut into small pieces with suitable shapes and <sup>415</sup> sizes. A gel solution that contained PVA powder  $(1.0 \text{ g})$  and  $H_3PO_4$  416 (1.0 g) in water (10.0 mL) was coated on top of the prepared films <sup>417</sup> and dried in air for 5 h. Then two such-prepared standing enokitake- 418 like nanowire-based gold film electrodes were assembled with <sup>419</sup> sandwiched electrolytes to form a symmetrical electrochemical 420 capacitor. 421

Wireless Facial Expression Monitoring. The circuit was 422 composed of nine standing enokitake-like nanowire-based gold film <sup>423</sup> sensors for measuring 11 facial muscle groups, and the supporting 424 circuit was constructed with 3.3 V power supply and 13 330  $\Omega$  425 resistors. After the standing enokitake-like nanowire-based gold film <sup>426</sup> sensors were mounted on the particularly targeted muscle groups on 427 the subject's face, electrical responses of each sensor were recorded. A 428 Bluetooth low energy technology was used to transfer the analogue 429 reading data of each sensor to an Android OS-equipped mobile device 430 (e.g., phone or pad style device). A specially designed app, already 431 installed on the mobile device, first went through a machine learning <sup>432</sup> session, which was referenced to the FACS library from Ekman's 433 group. The FACS contributes as the reference blueprint for pattern 434 recognitions to detect various facial expressions. This system was able 435 to process electrical responses from facial muscle groups in real time, 436 provided the baseline for measuring subject's detailed facial 437 movement, and eventually translated it to different emotional <sup>438</sup> expressions. The system was also able to create a data dictionary to 439 store the data based on the nine sensor readings to specific muscle <sup>440</sup> groups. 441

#### ASSOCIATED CONTENT 442

#### $\bullet$  Supporting Information  $443$

### The Supporting Information is available free of charge on the <sup>444</sup>

ACS Publications website at DOI: 10.1021/acsnano.8b05019. <sup>445</sup> [Movie](http://pubs.acs.org) [S1:](http://pubs.acs.org) [Thin](http://pubs.acs.org) [sta](http://pubs.acs.org)nding en[okitake-like](http://pubs.acs.org/doi/abs/10.1021/acsnano.8b05019) [nanowire](http://pubs.acs.org/doi/abs/10.1021/acsnano.8b05019) films  $_{446}$ 

on the back of a human hand, stretching and releasing <sub>447</sub>  $(AVI)$  448

[Movi](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_001.avi)e S2: Repeatable adhesion tape test  $(AVI)$  449 Movie S3: Finite element analysis model[ing](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_002.avi) of strain  $_{450}$ 

deformation for standing nanowire film (AVI) 451 Movie S4: Wireless facial expression m[onito](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_003.avi)ring from  $_{452}$ standing nanowire-based smart sensors (AVI) 453 Figures S1–S15, Notes S1 and S2, Ta[bles S](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_004.avi)1 and S2,  $_{454}$ 

and additional references ([PDF\)](http://pubs.acs.org/doi/suppl/10.1021/acsnano.8b05019/suppl_file/nn8b05019_si_005.pdf) 455

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464 Notes

<sup>465</sup> The authors declare no competing financial interest.

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