Paper-supported WS\textsubscript{2} strain gauges

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ABSTRACT

Environmentally friendly and low-cost sensors are needed for the next generation disposable electronics applications. Given its low-cost, availability and biodegradability, paper-based devices are a very promising. Here we demonstrate the fabrication of a tungsten disulphide (WS\textsubscript{2}) strain sensor on standard copy paper. The WS\textsubscript{2} is deposited through direct abrasion of WS\textsubscript{2} powder against the paper surface making the fabrication of the device low-tech and cost effective. The fabricated strain gauge devices present gauge factors up to ~70 for strains in the ± 0.5 % range. These values are ~9 times larger than that obtained on devices with the same geometry but using a graphite film instead a WS\textsubscript{2} as a sensitive material. We demonstrate the potential of these WS\textsubscript{2}-on-paper strain gauges by integrating them directly on a paper cantilever to sense mass and forces. We show how this very simple device can detect sub-milligram masses. Moreover, we also demonstrate the capability of transducing motion in mechanical resonators by gluing a WS\textsubscript{2}-on-paper strain gauge on their surface.

Keywords: paper electronics; strain gauge; sensor; low-cost electronics; van der Waals materials; tungsten disulfide WS\textsubscript{2}
Ubiquitous electronics applications like Internet of things, wearables or smart patches/tags will come together with an exponential growth of the electronic component production, eventually leading to an urgent problem of electronic waste handling.[1–3] In fact, only a very small fraction of standard silicon-based electronic components can be recycled thus motivating the exploration of alternative, eco-friendlier, substrate materials for electronic components. Although silicon will be still necessary in applications where a high level of electronic components integration is crucial, in other applications (like sensors) using alternative substrate materials could be easily achieved in the short term.

Among the different biodegradable substrate materials, paper has attracted the interest of the materials science community because of its ultra-low cost and availability.[4–7] Fabrication of electronic devices on paper substrates is, however, very challenging. In fact, paper is rough, fibrous and permeable which hampers the use of conventional lithographic techniques to fabricate electronic components. Up to now, most approaches are based on integrating electronic materials on paper substrates through inkjet printing.[8–13] Nonetheless, specialized paper surface treatments (that increase the price ~10 times and hamper its biodegradability) are required to print electronic components on paper. Recently, we demonstrated the integration of different van der Waals materials on standard printer/copy paper (untreated) through direct abrasion of powders of van der Waals materials against the rough surface of paper.[14–18] This deposition process is similar to drawing/writing with a pencil on paper: the friction force generated when a van der Waals material is rubbed against paper cleaves the crystals leading to a network of interconnected platelets.

Here, we demonstrate the fabrication of strain sensors on standard copy paper substrates using abrasion-induced deposited WS\textsubscript{2} films as sensing material. We obtained gauge factors up to ~70 for strains in the ± 0.5 % range, a factor 9-10 times larger than that
measured using graphite based paper-supported strain gauges. We illustrate the potential of these WS\(_2\)-on-paper strain gauges by fabricating a simple force/mass sensor and by transducing motion of mechanical resonators.

As shown in Figure 1a, the paper-supported strain sensor devices are fabricated by abrasion induced deposition of WS\(_2\) onto standard copy paper. [15–17] Briefly, the outline of the device channel and electrodes is printed out in a piece of standard copy paper (80 g/m\(^2\)) with an office laser printer. Then, Post-It tape is used to create a stencil to delimit the channel area and micronized WS\(_2\) powder (0.6 micron APS Micronised WS\(_2\), HAGEN Automation ltd.) is rubbed against the surface of copy paper with a cotton swab to deposit the channel. The continuity of the film is tested with a handheld multimeter and the rubbing process is repeated until reaching a resistance between two probes separated by ~1-2 mm in the ~1-5 M\(\Omega\) range. After that, the stencil is removed and graphite electrodes are deposited by direct drawing with a 4B pencil (~80% graphite content[19]) onto the WS\(_2\) and paper surfaces following the printed electrode outline contour.

Figure 1b shows a picture of one of the WS\(_2\) strain sensors fabricated on standard copy paper. A very comprehensive characterization of the morphology, chemical composition, electrical and optical properties of abrasion-induced deposited WS\(_2\) films on copy paper can be found in Ref. [17] and we address the reader to that work for further details. Figure 1c shows a scanning electron microscopy (SEM) image of the interface between the bare, uncovered, paper substrate and the deposited WS\(_2\) film. On the paper substrate one can readily resolve the fibrous structure, due to the cellulose fibers composing the substrate. The WS\(_2\) film covers the surface of the paper, filling in the gaps between the cellulose fibers, and creating a compact film of interconnected flakes.
Figure 1. WS₂-on-paper strain sensors. (a) Steps in the fabrication of the WS₂-on-paper strain sensors. (a.1) The outline of the device is printed on standard copy paper with an office laser printer. (a.2) The channel area is masked using Post-It tape. (a.3) WS₂ powder is rubbed against the unmasked paper with a cotton swab and the mask is removed (a.4). (a.5) Graphite electrodes are deposited onto the WS₂ channel by directly drawing with a 4B pencil. (a.6) Copper tape is attached to the graphite leads to allow soldering electrical connections. (b) Higher magnification picture of one device. (c) False coloured scanning electron microscopy image of the paper covered with WS₂-bare paper interface.

To characterize the suitability of these devices in strain sensing applications we load the fabricated devices into a home-built three-points bending jig apparatus (see the inset in Figure 2a).[20] The devices are subjected to well-defined strains while the electronic transport characteristics are determined. Figure 2a shows current vs. voltage characteristics (IVs hereafter) acquired at different levels of uniaxial strain, including tensile strain (positive strain values) and compressive strain (negative strain values). The resistance of the device changes monotonically when uniaxial strain is applied, as indicated by the change in slope of the IVs. For tensile strains the resistance increases while for compressive strains the resistance decreases. The overall dependence of the device resistance on the applied strain can be better resolved in Figure 2b where the relative change in resistance (with respect to the resistance at 0% strain, R₀) is displayed for different strain levels. The gauge factor (GF) or strain factor of the WS₂ paper-supported strain gauge device can be defined as:
GF = (ΔR/R₀)/ε

where ε is the strain and ΔR is the difference between the resistance when subjected to a certain strain and R₀. The gauge factor is a very suitable figure-of-merit that allows to directly compare the performance of different strain sensors. In the WS₂-on-paper strain gauge shown in Figure 2b we obtain a gauge factor of ~60 for tensile strain and ~40 for compressive strain. For comparison (Table 1), state-of-the-art metallic thin-film based strain gauges have gauge factors in the 2-10 range.[21] Recently, other paper-supported strain sensors, based on black phosphorus, ITO inks and carbon black, have been reported with gauge factors in the 4-40 range.[22–24] Graphite and graphene strain sensors on paper have been also demonstrated but the gauge factors span over a very broad range of values, 3 to 800.[25–28] The reason for such a large variation in gauge factor of graphite/graphene based strain sensors in different reports is unclear. We thus decided to carry out a benchmarking test with graphite-based strain sensors fabricated and tested with the exact same protocol as the WS₂ sensors. Figure 2c shows the measured relative change in device resistance as a function of strain, measured for 17 WS₂ and 12 graphite (4B pencil, ~80% graphite content[19]) devices. One can observe that the WS₂ devices show a stronger resistance change upon straining as compared with the graphite devices. The Supporting Information shows datasets for 5 different WS₂ devices and one graphite (4B) device for comparison. We have also tested graphite devices fabricated out of pure nano-graphite powder (MKN-CG-50, Lower Friction, MK IMPEX CORP) obtaining a similar behaviour than that of 4B pencil based devices, we address the reader to the Supporting Information Figure S3 to see the results. From the slope at low strain values one can quantitatively extract the gauge factor of the different devices. Figure 2d presents the statistical summary of the results in a box-plot. Our graphite devices show gauge factors with a median value of 5.7 (for tensile strain) that are compatible with the lower
bound of the reported values in the literature for graphite/graphene strain sensors on paper. On the other hand, WS$_2$-on-paper devices show gauge factor values up to 67.6, ~9-10 times larger than that of graphite.

Table 1. Summary of gauge factor values reported for other conventional and paper-supported strain gauges. The table indicates the strain sensitive material used, the substrate, the method to deposit the sensing material and the reported gauge factor. Gauge factor values highlighted with $^t$ or $^c$ indicate values obtained upon tensile or compressive strains, respectively.

<table>
<thead>
<tr>
<th>Sensing material</th>
<th>Substrate</th>
<th>Method</th>
<th>Gauge factor</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metallic thin-film</td>
<td>Flexible polymer (usually Kapton)</td>
<td>Lithography + metal deposition</td>
<td>2-10</td>
<td>[21]</td>
</tr>
<tr>
<td>Black phosphorus</td>
<td>Paper</td>
<td>Sonochemistry, hydrothermal</td>
<td>6.1</td>
<td>[22]</td>
</tr>
<tr>
<td>Carbon black</td>
<td>Paper</td>
<td>Dip-coating</td>
<td>4.3</td>
<td>[23]</td>
</tr>
<tr>
<td>ITO nanoparticles</td>
<td>Paper</td>
<td>Hand-painting</td>
<td>41.98 $^t$; 21.36 $^c$</td>
<td>[24]</td>
</tr>
<tr>
<td>Graphite glue</td>
<td>Paper</td>
<td>Stencil printing</td>
<td>804.9 $^t$; 142.1 $^c$</td>
<td>[25]</td>
</tr>
<tr>
<td>Water-based graphene inks</td>
<td>Paper</td>
<td>Inkjet-printing</td>
<td>125</td>
<td>[26]</td>
</tr>
<tr>
<td>Graphite pencil-trace (HB)</td>
<td>Paper</td>
<td>Drawing</td>
<td>26</td>
<td>[27]</td>
</tr>
<tr>
<td>Graphite pencil-trace (2B)</td>
<td>Paper</td>
<td>Drawing</td>
<td>536.6</td>
<td>[28]</td>
</tr>
<tr>
<td>Graphite pencil-trace (2B)</td>
<td>Paper</td>
<td>Drawing</td>
<td>34</td>
<td>[29]</td>
</tr>
<tr>
<td>Carbon black/carbon nanotube (CB/CNT)</td>
<td>Paper</td>
<td>Dip-coating</td>
<td>7.5 $^t$; 2.6 $^c$</td>
<td>[30]</td>
</tr>
<tr>
<td>Molybdenum carbide-graphene (MCG) composites</td>
<td>Paper</td>
<td>Direct laser writing</td>
<td>73 $^t$; 43 $^c$</td>
<td>[31]</td>
</tr>
<tr>
<td>Reduced graphene oxide (rGO)</td>
<td>Paper</td>
<td>Drop-casting</td>
<td>66.6 ± 5</td>
<td>[33]</td>
</tr>
<tr>
<td>Graphite</td>
<td>Hybrid graphite-paper</td>
<td>Preparation of graphite infiltrated paper</td>
<td>27</td>
<td>[32]</td>
</tr>
<tr>
<td>Graphene</td>
<td>Mulberry paper</td>
<td>Meyer-rod coating</td>
<td>3.82</td>
<td>[34]</td>
</tr>
<tr>
<td>Carbon paper</td>
<td>Carbon paper</td>
<td>Pyrolysis</td>
<td>25.3</td>
<td>[35]</td>
</tr>
<tr>
<td>Carbonized crepe paper</td>
<td>Carbonized crepe paper</td>
<td>Pyrolysis</td>
<td>10.10</td>
<td>[36]</td>
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<tr>
<td>Laser-fabricated graphitic platelets</td>
<td>Polymide (PI)/paper</td>
<td>Direct laser writing</td>
<td>13</td>
<td>[37]</td>
</tr>
<tr>
<td>Au nanoparticles</td>
<td>Abrasive paper</td>
<td>Direct-current (DC) sputtering</td>
<td>75.8 $^t$; 10.7 $^c$</td>
<td>[38]</td>
</tr>
<tr>
<td>Single-layer graphene</td>
<td>PDMS</td>
<td>Chemical vapor deposition (CVD), photolithography</td>
<td>42.2</td>
<td>[39]</td>
</tr>
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<td>WS$_2$</td>
<td>PET</td>
<td>Atomized spray casting deposition</td>
<td>14</td>
<td>[40]</td>
</tr>
<tr>
<td>WS$_2$</td>
<td>Paper</td>
<td>Direct abrasion</td>
<td>67.6 $^t$; 51.0 $^c$</td>
<td>This work</td>
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<tr>
<td>Graphite (4B)</td>
<td>Paper</td>
<td>Direct abrasion</td>
<td>10.2 $^t$; 7.8 $^c$</td>
<td>This work</td>
</tr>
<tr>
<td>Graphite (nanographite)</td>
<td>Paper</td>
<td>Direct abrasion</td>
<td>9.5 $^t$; 11.1 $^c$</td>
<td>This work</td>
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</tbody>
</table>
Figure 2. Strain dependent resistance of WS$_2$-on-paper devices. (a) Current vs. voltage characteristics of a paper supported WS$_2$ strain gauge device, measured for different uniaxial strains. (Inset) Picture of the device loaded in a three-point bending jig apparatus to apply controllable strains. (b) Resistance changes as a function of the applied strain. The gauge factor (GF) can be extracted from the slope of the curve. (c) Comparison between the strain dependent resistance changes measured on 17 WS$_2$ devices and that measured on 12 devices with graphite sensing channel. (d) Box plot summarizing the gauge factors measured, within ± 0.2% strain range, for the WS$_2$- and graphite-based devices.

In order to illustrate the potential of the WS$_2$-on-paper strain sensors we have fabricated paper-based force and mass sensors that use a WS$_2$ strain gauge as transducer. The force/mass sensor is based on a cantilever made of copy paper and we fabricated a WS$_2$
strain gauge on its base to convert the deflections of the cantilever into an electrical signal (see Figure 3a). We first test the use of such device to sense mass by loading the cantilever free end with test-masses and measuring the change in resistance (Figure 3b). Upon mass loading, the cantilever deflects downwards (see insets in Figure 3b) creating a tensile strain on the base of the cantilever that can be detected with the WS₂ strain gauge integrated directly on the surface of the paper cantilever. The resistance of the device raises gradually with increasing mass loading, and one can obtain a slope of ~160 Ω/mg that represents the mass sensitivity of the sensor. In order to test the reproducibility of this sensor we have subjected it to 80 deflection cycles by gluing a magnet at the free end of the cantilever and mounting another magnet on a motorized stage to deflect the cantilever through magnetic force repulsion. Figure 3c presents the resistance changes during these deflection cycles, showing how the device responds fast to the deflection change and with a good reproducibility, with an overall drift of less than 0.8% in the 5 min measurement. An alternative way of sensing mass with this device is to use the WS₂ strain gauge to study the dynamics of the paper cantilever. Figure 3d shows the resistance of the WS₂ film as a function of time for a cantilever loaded with 1 to 4 test masses (73.6 mg each test mass) when it is mechanically excited. One can observe that the current oscillates in time and its amplitude decays, as expected for a damped harmonic oscillator. One can fit the measured resistance vs. time traces to a damped oscillator to accurately extract the resonance frequency of the paper cantilever, that decreases monotonically upon mass loading (see Figure 3e). Therefore, one can determine the mass load alternatively to the direct resistance change measurement from the determination of the resonance frequency of the cantilever. For low mass load, the change in frequency is in the order of 20 mHz/mg and thus one could easily resolve sub-mg test masses.
Figure 3. Paper-based cantilever with integrated WS$_2$ strain gauge. (a) Picture of the paper cantilever with a WS$_2$ film strain gauge integrated at the base of the cantilever. (b) Resistance of the strain gauge as a function of the mass load. (Insets) Pictures of the cantilever: unload (top left) and loaded with ~1.0 g (bottom right). (c) Resistance of the WS$_2$ film vs. time while the paper cantilever is subjected to ~80 cycles of bending/releasing. (d) Resistance of the WS$_2$ film vs. time acquired when the paper cantilever, loaded with different test masses, is mechanically excited to induce a ring-down. The datapoints have been fitted...
to a damped harmonic oscillator model to extract the resonance frequency of the cantilever. (e) Resonance frequency of the cantilever measured for different mass loading conditions.

The capability of the WS$_2$-on-paper strain gauges to transduce strain dynamically (Figure 3e) motivated us to test the performance of these devices to transduce the motion of mechanical resonators by directly sticking a paper with a WS$_2$ strain gauge on the surface of the mechanical resonator. Figure 4a shows pictures of a WS$_2$-on-paper strain sensor glued on the backside of a piezo-buzzer disc. The piezo-buzzer is then excited by sending a sine wave signal to the piezoelectric material with a function waveform generator (Tenma 72-14110) and the resistance of the WS$_2$-on-paper strain gauge is measured. Figure 4b shows the resistance of the WS$_2$ strain gauge as a function of time when the piezo-buzzer is excited with sine waves of frequencies 0.5 Hz, 1 Hz, 2 Hz and 4 Hz. One can see how the resistance of the WS$_2$ strain gauge oscillates following a sine wave of the same frequency as the piezo-buzzer excitation, indicating that the motion of the piezo-buzzer induces a strain on the WS$_2$-on-paper that is transduced as a change in resistance. By repeating the experiment at different piezo-buzzer excitation frequencies one can get information about the mechanical resonances of the piezo-buzzer disc (Figure 4c). In this example the disc has a broad resonance around 256 Hz. Interestingly, the paper-based strain gauge could be used to transduce the motion of the piezo-buzzer up to 2 kHz (the upper frequency limitation of our electrical measurement system) and thus the response time of the WS$_2$-on-paper strain gauge is < 0.5 ms.
Figure 4. Use of WS$_2$ strain gauge as a motion transducer. (a) Pictures of a piezoelectric disc buzzer (top) with a WS$_2$-on-paper strain gauge glued on its back side (bottom). (b) Resistance of the WS$_2$ film vs. time measured when the piezoelectric buzzer is excited with a sine wave of 0.5 Hz, 1 Hz, 2 Hz and 4 Hz (10 V$_{pp}$), respectively. (c) FFT amplitudes at various excitation frequencies.

CONCLUSIONS

In summary, we fabricated strain sensors on standard copy paper substrates integrating a WS$_2$ film, as sensing material, contacted with graphite electrodes. The WS$_2$ was deposited through a low tech and cost effective all-dry abrasion-induced deposition method that does not require of any specialized fabrication instrumentation. The graphite electrodes were directly drawn onto the WS$_2$ film using a high-graphite content pencil (B-type). The resistance of the strain sensors strongly depends on the applied strain, reaching gauge factor values up to $\sim$70, 9-10 times larger than the values obtained when a graphite film is used. We demonstrate the direct integration of the WS$_2$-on-paper strain sensor on a
paper cantilever, allowing for the transduction of cantilever deflection, allowing for mass and force sensing. We demonstrate a mass sensitivity of ~160 Ω/mg using this simple disposable mass sensor. We have also demonstrated how gluing these paper-based sensors to a mechanical resonator we can transduce its motion even at frequencies > 2 kHz. The results presented here open the door to the integration of other van der Waals materials in the fabrication of biodegradable, low-cost and disposable paper-supported strain sensors.

MATERIALS AND METHODS

Materials: Standard (untreated) copy printer paper (80 g/m²) were used as supporting substrates because their low cost and availability. Tungsten disulfide (WS₂) from HAGEN automation Ltd. (0.6 micron APS Ultra Grade Micronised) was used as sensing channel material. Graphite pencil (4B, Faber Castell) was used to pattern graphite-based electrical leads to connect the WS₂ channel to the readout electronics. Nano-graphite powder with the average particle size of 50 nm (PN: MKN-CG-50) was purchased at Lowerfriction Lubricants.

SEM and EDX: Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX), using a FE-SEM, FEI Nova NANOSEM 230, was used to characterize the morphology and the composition of the WS₂ films deposited on paper. An electron energy of 7 keV were used for imaging and 14 keV for EDX spectroscopy.

Electrical measurements: Electrical measurements were carried out with a Keithley 2450 source measure unit. The WS₂-on-paper devices were fixed on a home-made three-points bending setup to conduct the well-defined tension and compression deformations. The electrical resistance of the device was determined by measuring current vs. voltage.
characteristics at various uniaxial strains. The variation of electrical resistance of the strain sensor was monitored with a fixed voltage of 1 V.

**Supporting Information:** Supplementary Information includes: extra datasets of some WS$_2$-on-paper strain gauge devices, electrical characteristics of a pencil-on-paper strain gauge upon strain, electrical characteristics of several nanographite-on-paper strain gauge upon strain.

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