Ultrafast humidity sensor based on liquid phase exfoliated graphene

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Abstract
Humidity sensing is important to a variety of technologies and industries, ranging from environmental and industrial monitoring to medical applications. Although humidity sensors abound, few available solutions are thin, transparent, compatible with large-area sensor production and flexible, and almost none are fast enough to perform human respiration monitoring through breath detection or real-time finger proximity monitoring via skin humidity sensing. This work describes chemiresistive graphene-based humidity sensors produced in few steps with facile liquid phase exfoliation followed by Langmuir–Blodgett assembly that enables active areas of practically any size. The graphene sensors provide a unique mix of performance parameters, exhibiting resistance changes up to 10% with varying humidity, linear performance over relative humidity (RH) levels between 8% and 95%, weak response to other constituents of air, flexibility, transparency of nearly 80%, and response times of 30 ms. The fast response to humidity is shown to be useful for respiration monitoring and real-time finger proximity detection, with potential applications in flexible touchless interactive panels.

Supplementary material for this article is available online

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(Some figures may appear in colour only in the online journal)

1. Introduction
Humidity monitoring is essential for numerous applications across industries, such as environmental and industrial monitoring, and healthcare [1, 2]. Aside from traditional uses in monitoring atmospheric and room conditions, technological progress keeps enabling new uses for humidity sensors. In healthcare, humidity sensing could be used for human respiration monitoring [3, 4] due to the high level of water vapor in breath. In electronics and robotics industries, humidity clouds near human skin could be used to detect finger position for touchless control interfaces [5, 6]. However, humidity sensors made of established materials such as metal/polymer composites have temporal response rates that are far too slow for these applications, in the range 5–50 s [2]. Emerging composite and nanomaterials such as ZnO and Pd–SnO2 also suffer from the same debilitating disadvantage [7, 8]. More recently, graphene and graphene oxide have emerged as a promising material for fast humidity sensing, with response times ranging from 10’s of milliseconds to a few seconds, depending on the method of production [9–15].

Compared to many other materials, graphene has the added benefits of being thin, flexible, and transparent, enabling applications in wearable and flexible electronics. Nevertheless, graphene humidity sensors to date have been made from graphene that is either industrially irrelevant (mechanically exfoliated), expensive (chemical vapor deposited, CVD) or...
made with several complex chemistry steps, such as with reduction of graphene oxide.

Here, we demonstrate fast humidity sensors made with an inexpensive and facile production method that is compatible with large-area sensor production. The active sensing area is made from liquid-phase exfoliated graphene [16] that requires only a single ultrasonic processing step. The humidity response times of our sensors are as low as \(~\sim 30\) ms, which allows us to show real-time breath monitoring and finger proximity detection as exemplary applications of ultrafast humidity sensing. Breath monitoring and proximity detection are only two examples of potentially numerous applications of these novel ultrafast humidity sensors, enabled by the fast response. We demonstrate sensing capability on three different substrates, including flexible transparent ones. The sensors fare very well against standard gas sensor performance metrics, such as insensitivity to other components of air and response time [17].

2. Material and methods

2.1. Fabrication of graphene films and humidity sensors

The graphene dispersion was produced by dissolving graphite powder (Sigma Aldrich, product no. 332461) at a concentration of 18 mg ml\(^{-1}\) in N-Methyl-2-pyrrolidone (NMP) (Sigma Aldrich, product no. 328634). The dispersion was sonicated in a low energy ultrasonic bath for 14 h. After sonication the dispersion was centrifuged at 3000 rpm for 60 min in order to separate non-exfoliated graphite flakes, which remain in the precipitate, and the exfoliated graphene flakes which are dispersed in the supernatant. A small volume of the supernatant is added to deionized water (18 M\(\Omega\) cm\(^{-1}\)) resulting in self-assembly of graphene nanoplatelets into a thin film on the water/air interface. The thin film is deposited on a pre-immersed substrate of choice following the Langmuir–Blodgett method [16, 18–20].

Our film, made of graphene that is exfoliated in the liquid phase and assembled with the Langmuir–Blodgett method, consists of graphene nanoplatelets in contact with each other, as shown in the atomic force micrograph in supporting information (figure S1) (available online at http://stacks.iop.org/NANO/31/025505/mmedia).

Although nanoplatelets conform to a distribution of thicknesses, the average thickness of the film is \(~\sim 10\) layers of graphene (3.4 nm), as measured with UV–VIS absorption spectroscopy and shown in supporting information (figure S2). For well-defined channel geometry and accurate sheet resistance measurements, we use a SiO\(_2\)/Si substrate with four microfabricated metal contacts (figure 1(a)). The substrate is a 380 \(\mu\)m thick n-doped Si wafer with a thermally grown 800 nm thick SiO\(_2\) insulating layer. We deposit a layer of chromium (20 nm) and a layer of gold (100 nm) with radio-frequency cathode sputtering. Subsequently the layers of chromium and gold are coated with 0.5 \(\mu\)m of photoresist (AZ-1505) that is subjected to direct laser writing (LW405, MicroTech, Italy) [21] to pattern the contacts. The gold is removed with a solution of potassium iodide and the chromium is removed with a solution of ammonium cerium (IV) sulfate. The wafer is diced into 3 \(\times\) 3 mm chips, each chip containing a set of four metal contacts. After film deposition, the chips are mounted to TO-8 housing. Sensor fabrication is reproducible, with AFM and UV–VIS measurements yielding the same average thickness for every device made.

The inset of figure 1(a) depicts an optical micrograph of the graphene film deposited on the contacts. Darker spots indicate remaining unexfoliated or thick graphite material. The film area between contacts has dimensions of \(~\sim 1500 \mu\text{m} \times 190 \mu\text{m}\). Taking into account film geometry yields sheet resistance of 3–7 k\(\Omega\) sq\(^{-1}\) for our films consistently, the smallest values reported for post-processing-free single-deposition Langmuir–Blodgett graphene films to date [16, 18, 20, 22]. Such small sheet resistance is a result of fabrication process streamlining and careful four-terminal resistance measurements. For obtaining the largest sensing area and highest signal-to-noise ratio, we employ a commercial ceramic substrate with pre-made interdigitated electrodes (DropSens IDEAU200), figure 1(b). This substrate is easy to handle, versatile, low-cost, and easily connected to macroscopic wires by soldering, and the active sensing area is \(~\sim 15\) mm\(^2\). Finally, for flexible transparent humidity sensors we employ a polyethylene terephthalate (PET) substrate with macroscopic gold contacts thermally evaporated over a shadow mask, as in
Figure 2. Experimental setup for measuring humidity response. A homebuilt humidity chamber is designed with slots for the graphene sensor and a reference sensor. It has inlets for water vapor from a room humidifier and for purging with nitrogen $N_2$ gas. The sensors are connected to measurement electronics.

2.2. Humidity sensing

We perform humidity sensing in a homebuilt humidity chamber made of polytetrafluoroethylene (PTFE), as depicted in figure 2. The chamber is equipped with separate valves for injection of water vapor and another gas, a gas outlet valve, and auxiliary connectors. Our sensor in its TO-8 housing is integrated into a custom-made PTFE plug that we insert into a matching slot in the chamber, next to a reference humidity sensor (Honeywell HIH-4000-001). A thermocouple is placed near the sensors to measure the local temperature. All measurements were performed at room temperature ($21 \degree C$–$23 \degree C$). We apply a current of $10 \mu A$ between the outer electrode pair and measure the induced voltage across the inner electrode pair to obtain the resistance. The voltage was measured with a Keysight 34 461 a DMM. In the cases of the commercial ceramic substrate and the PET substrate we monitored two-terminal resistance with the same DMM operated in ohmmeter mode. Relative humidity (RH) was decreased to 8% by purging the sealed chamber with nitrogen ($N_2$) gas. Water vapor is produced with a commercial room humidifier and injected into the chamber at a constant flow rate. Once a desired humidity level is reached the water vapor inlet valve is manually closed.

To measure sensor speed below the limit imposed by humidity chamber filling time, we placed the sensors on a table in free space at RH $\sim$40%. A nitrogen gun was used to induce nitrogen flow across the sensor surface, quickly drying the active area while we monitored the sensor response and recovery times [11].

3. Results and discussion

3.1. Humidity sensing

The humidity sensing performance of our devices was tested by monitoring device resistance while controlling relative humidity in the humidity chamber. Figure 3(a) depicts three cycles of a humidity ramp in the test chamber and corresponding measurements with our graphene sensor on the ceramic substrate with interdigitated electrodes (black) and the reference sensor (purple). During each cycle the humidity in the chamber is increased from $\sim$8% to 95% and then decreased to $\sim$8%. The resistance of our sensor rises with humidity, as was shown earlier for other films that consist of conducting NbS$_2$ nanoplatelets [23]. The increase in resistance is attributed to water adsorption at nanoplatelet edges and between the platelets, both of which disrupt electrical conduction paths of the film. In the case of graphene, other mechanisms could also play a role, such as electron donation from graphene to water and disruption of molecular symmetries of graphene by the water molecules [24]. For a clear perception of the sensitivity of our sensor, we plot a second vertical axis next to the resistance axis that depicts the percent change of resistance, described by $S = 100 \cdot \frac{\Delta R}{R_0}$, where $R_0$ is the initial resistance value and $\Delta R$ is the difference between the given and the
Figure 3. Response to humidity and other constituents of air. Measurements are conducted on the ceramic substrate with interdigitated electrodes. (a) Graphene sensor response measured in conjunction with the reference sensor response over three cycles of ramping RH from ~8% to 95%. (b) Graphene sensor response to repeat stepwise increase of relative humidity, from 26% to 88%, in time. (c) Peak sensor response as function of maximum RH. (d) Sensor response to nitrogen, oxygen and water vapor over 60 s each. (e) Response time of graphene sensor when a nitrogen N$_2$ gun is used to flush the device. (f) Recovery time of graphene sensor after flushing with nitrogen N$_2$ gun.

initial resistance value. The resistance of the graphene sensor changes by 5% when changing humidity from 8% to 95%. The sensitivity is thus higher than reported for CVD graphene [10, 11], which is more costly than liquid phase exfoliation as per word graphene, and higher than for industrially irrelevant mechanically exfoliated graphene [25]. Sensor repeatability indicates that the likely dominant mechanism is physisorption of water molecules, with little chemisorption. There is a small baseline drift that occurs when the sensor rests in air which is visible on figure 3(a). A similar drift was observed in mechanically exfoliated bilayer graphene [11]. Although such a drift could potentially be detrimental to the practical use of graphene-based gas sensors, we found that the drift is fully reversible with heating to temperatures around 150 °C that could easily be achieved with an on-chip integrated heater [26].

Figure 3(b) depicts the response of the graphene sensor over several cycles with different maximum RH. In this image the baseline drift has been corrected for by linear subtraction. The raw data that includes the drift is provided in supporting information (figure S3). The relative change in resistance, S, as a function of maximum humidity is shown in figure 3(c). The sensor response is clearly linear with humidity ($r = 0.981$), which indicates potential for applications in
Figure 4. Respiration monitoring. (a) Monitoring fast, normal and deep breathing with graphene sensor on ceramic substrate. (b) Monitoring breathing rate on flat PET substrate, and (c) on PET that is bent at an angle of 10 degrees, as seen in (d).

diverse conditions. In the case of our graphene sensors on Si/SiO$_2$ substrates, the linearity is similar to that reported here, while the sensitivity is $\sim$10 times smaller (see supporting information, figure S4). The sensing response on the two substrates should not be directly compared, because of the vastly different active sensing areas and contact spacing. The ceramic substrate is optimized for high sensing performance, whereas the contact design on the Si/SiO$_2$ substrate is optimized for sheet resistance measurements.

To confirm that our sensor reacts to water vapor and not to other constituent gases of air, we tested the response to nitrogen and oxygen. Figure 3(d) depicts the sensor response to nitrogen gas (injected into the chamber at t = 0 s), to oxygen gas replacing nitrogen (at t = 60 s), and finally to water vapor injected instead of oxygen (at t = 120 s). The sensor does not respond to nitrogen N$_2$, reacts very little to O$_2$ and has a strong response to H$_2$O gas, an effect that could be used to implement selectivity. Humidity was reduced to $\sim$0% before starting the experiment.

Sensor response and recovery times cannot be measured in the humidity chamber due to the limited chamber-filling and chamber-flushing times, hence we proceed to measure the sensor response time by rapidly drying the sensor surface in ambient with a nitrogen gun and observing sensor dynamics. In figure 3(e) we show nitrogen gun drying of the sample, with observed rapid recovery shown in figure 3(f). We set a 10% and a 90% change threshold for measuring rise and fall times. The sensor responds in 28 ms and recovers in 30 ms. Similar dynamics are observed in other samples on the same substrate. The obtained response is significantly faster than that reported earlier for single-layer and double-layer CVD graphene ($\sim$700 ms) [10, 11] and is two orders of magnitude faster than the commercial reference sensor (Honeywell HIH6100 Series Datasheet). The response time in the case of our graphene sensors on Si/SiO$_2$ substrates is longer, in the range of 240 ms (see supporting information figure S5). The fast response of our sensors is likely due to additional carrier scattering introduced by edge defects [27, 28].

3.2. Respiration monitoring

High-speed humidity sensors enable the monitoring of human respiration by breath detection. To test the usefulness of our sensors for respiration monitoring, we placed the sensors on a table and had a volunteer breathe near the sensor surface. Figure 4(a) depicts the response of a sensor on a ceramic substrate to breathing cycles in a fast, regular, and slow pattern. A recording of respiration detection is shown in Supporting Video 1. It is evident that the sensor responds to human respiration and can be used as a biometric detector of respiration rate. In real-life conditions outside the humidity chamber, the resistance changes by up to 20% during breathing, which offers an excellent on/off ratio that is useful for applications. The sensitivity to breath is larger than can be found in literature that describes other graphene-based sensors, which are also generally produced with more complex chemistry steps.
Figure 5. Finger proximity detection. (a) Optical image of the touchless proximity sensing experiment. (b) Resistance as a function of fingertip distance to device. (c) Demonstration of ultrafast proximity detection as a finger is swept across the device at different distances. (d) Response of touchless sensor to metallic tweezers and hand with glove.

The high sensitivity of our sensor is likely due to the abundance of reactive edge sites in the film consisting of interconnected nanoplatelets. The sensor on the SiO$_2$/Si substrate also responds to breath, although with a smaller sensitivity (see supporting information figure S6). A resistance change of $\sim$24% is measured for respiration monitoring on the ceramic substrate (figure 4(a)), while changes of $\sim$6%–8% are measured on the PET substrate (figures 4(b) and (c)).

3.3. Transparent flexible sensors

For certain applications, such as for monitoring the respiration rate of first responders or medical patients via sensors attached to transparent masks, or for transparent touchless control panels, it would be advantageous to have the sensor on a transparent substrate. Results of respiration monitoring of our large-area sensors on PET are shown in figure 4(b). Qualitatively, the performance is similar as on the rigid substrate. Quantitatively, the response is an order of magnitude weaker on PET, with a more pronounced background drift. We emphasize again that the sensing performance on the different substrates should not be directly compared, as the contact and sensor geometry is differently optimized on the various substrates for different purposes. The speed of sensor response on a PET substrate is $\sim$20 ms, as shown in supporting information (figure S7). We purposely made the sensor on a flexible substrate to demonstrate compatibility with flexible electronics and wearable technology. Flexing the substrate is not detrimental to the humidity sensing performance of the sensor, and results in similar response to breath, as seen in figure 4(c). The data shown in figure 4(c) was taken for a sensor bent with a curvature of 10 degrees, as in figure 4(d). The method of measuring curvature is presented in supporting information (figure S8). To demonstrate operational performance of our sensors under different conditions, we present sensor response at different temperatures (figure S9) and in a flexed state (figure S10). Sensitivity decreases with temperature, as is common for chemiresistive humidity sensors [30].

3.4. Finger proximity detection

Our devices have an interesting application in proximity sensing, as part of positioning interfaces for touchless screens and applications in robotics [5]. It is well known that human skin emits a cloud of moisture that decays over a distance of $\sim$1 cm, an effect that has been proposed as a working mechanism for positioning interfaces that detect the presence of a human finger [6]. However, practical realization of such interfaces has been elusive, primarily due to the low speed of the materials considered thus far. Devices based on VS$_2$ [6] and graphene oxide [31–33] have response and recovery times ranging from 1 s to more than 20 s, which causes a delay in finger position detection that is impractical. Figure 5(a) depicts an
optical image of the proximity detection experiment. A finger is held at a specific distance from the device as the resistance is measured. Figure 5(b) depicts the distance-dependent device response to the presence of a fingertip. The sensor responds at a finger distance of 10 mm and resistance significantly increases for smaller distances. To demonstrate ultrafast performance of our proximity sensor, the volunteer swipes his finger above the device at different distances (figure 5(c)). It is clear that the device responds to finger motion in real time, enabling practical development of novel man-machine interactive systems. Real-time finger proximity detection is demonstrated in Supporting Video 2. We show that the response to human finger proximity is due to humidity and not a capacitive effect by testing the sensor response to the presence of metal tweezers and a finger covered with a rubber glove (figure 5(d)). The maximum measured resistance change for an approaching finger is \(\sim 1.6\%\).

4. Conclusions

We have demonstrated humidity sensors based on graphene that are sensitive, thin, flexible, nearly 80% transparent, only weakly reactive to other constituents of air, and fast enough to be used for advanced applications such as respiration rate monitoring and finger proximity detection. The principles of operation shown here, combined with the ease of manufacture of the sensors, indicate strong technological potential for wearable health monitoring and touchless control panels. The demonstrated behavior is unparalleled in literature, surpassing other state of the art solutions in terms of sensor response and recovery times, ease of manufacture, substrate compatibility, transparency and scale-up potential.

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