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A novel hydrogen sensor based on Pt/WO$_3$/Si MIS Schottky diode

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I. SUMMARY
In this work, we investigate the static and dynamic gas response of Schottky diode based hydrogen sensor employing a Pt/WO$_3$/n-type Si configuration. The role and importance of tungsten trioxide as an insulating layer within the device is discussed with respect to the measured electronic properties. The WO$_3$ thin films were deposited using RF reactive magnetron sputtering. The surface morphology was studied by an atomic force microscopy (AFM) and the scan results indicated a smooth film with a roughness of 0.18 Å. From the X-ray photoelectron spectroscopy (XPS) characterization, it can be confirmed that the films were stoichiometric WO$_3$ with a thickness of about 4 nm (as measured by an ellipsometer). The $I$-$V$ characteristics and dynamic response with respect to H$_2$ gas were measured at elevated temperatures from 50 °C to 150 °C and the results indicate that the H$_2$ sensitivity of this device can exceed approximately 1000 % with an average response time of less than 10 seconds. We discuss and explain these observations in terms of current transportation mechanisms using the thermionic emission model and the change in the Schottky barrier height.

II. MOTIVATION AND RESULTS
With the increasing demand for cleaner and more efficient energy generation technology, hydrogen is now considered as one of the most promising candidates to address the issues of dwindling fossil fuel reservation. To build a hydrogen economy, one of the key elements is the ability to perform rapid and accurate concentration measurement of this substance as a fuel. In addition, detection for hydrogen leakage is essential to H$_2$ production, storage, transportation and usage due to its highly flammable and volatile nature [1].

Sensors based on semiconducting metal oxides have arisen as one of the most attractive choices for industrial and commercial applications for their simplicity and lightweight characteristics [2]. WO$_3$, a n-type semiconductor with a cubic ReO$_3$ structure, has an oxidation state of +6. The free electrons generated primarily from oxygen vacancies can serve as charge traps for sensing application. Hence, we can expect its superior sensing properties towards hydrogen gas than other oxides [3]. Furthermore, sensors fabricated with a two-dimensional WO$_3$ thin film can exhibit higher sensitivity, faster response and lower working temperature than their thick-film counterparts due to the quantum confinement of the heterostructure and strong polarization at the interface [4].

In this paper, we employ a 4 nm WO$_3$ thin-film insulating layer formed by RF-sputtering on n-type <100> silicon substrates. Fig. 1 shows the AFM micrograph of the deposited WO$_3$ film. The graph indicates that the surface is inhomogeneous and made of tiny grains and voids. From the XPS data, two doublet peaks that are associated with the W$_4$f$^{12}_{1/2}$ and W$_4$f$^{7/2}_{1/2}$ can be seen, confirming the presence of W$^{6+}$ in the WO$_3$ stoichiometric film. The $I$-$V$ characteristics of the sensor at 50 °C, 100 °C and 150 °C in the presence of synthetic air and 1 % H$_2$ in air are shown in Fig. 2. The inset of Fig. 2 shows a plot of the sensitivity, defined as $(I_{H_2} - I_{air})/I_{air}$, and it was found to decrease with increasing temperature. We explain this observation by the change in Schottky barrier at the interface of Pt and WO$_3$ thin film, as shown in Fig. 3. The dynamic response of 0.1 % H$_2$ in N$_2$ at temperatures ranging from 25 °C to 150 °C under 2.5 V bias is shown in Fig. 4. Results show that the sensor can achieve steady state with the response and recovery time in the order of seconds. Thus, we can conclude that by implementing a 4 nm WO$_3$ thin film layer into a Schottky diode type sensor, the sensitivity of the device can be significantly increased, and we have shown that these characteristics are suitable for sensing applications requiring fast response.

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REFERENCES


Fig. 1. (a) AFM micrograph (500 nm x 500 nm) (b) O1s and W4f XPS spectrum of the WO$_3$ thin film annealed at 500 °C for a minute.

Fig. 2. I-V characteristics of the WO$_3$ thin film sensor when exposed to Air and 1% H$_2$ in Air at 50 °C, 100 °C, 150 °C (insert table shows the H$_2$ sensitivity at 2 V).

Fig. 3. Schottky Barrier Height (SBH) in Air and SBH change when exposed to 1% H$_2$ in Air.

Fig. 4. 0.1% H$_2$ transient response in an ambient of pure N$_2$ at 25 °C, 50 °C, 100 °C, 150 °C with forward bias voltage of 2.5 V.