Hydrogen gas sensing properties of Pd/ZnO thin films grown on *n*-Si<100> substrates at room-temperature by thermal evaporation and sol-gel techniques: A comparative study

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The present paper compares the room temperature hydrogen (H_2) gas sensing properties of two Pd/nanocrystalline-ZnO thin film based Schottky contacts grown on *n*-type silicon (100) substrates by thermal evaporation and sol-gel techniques. The structural, surface and optical properties of the ZnO thin films under consideration are also presented. The Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectroscopy (EDS) measurements have been carried out to study the surface morphologies and elementary compositions of the ZnO films, respectively. The X-ray diffraction (XRD) analysis shows that the ZnO thin films grown by both the methods are polycrystalline in nature with a hexagonal wurtzite structure. The optical band gaps of the films are estimated from the photoluminescence (PL) spectroscopy as 3.26 eV and 3.28 eV for thermal evaporation and sol-gel techniques, respectively. The current–voltage (*I-V*) measurements have been carried out to study the electrical and hydrogen (H_2) sensing characteristics of Pd/ZnO Schottky contacts fabricated on the ZnO thin films grown by the two methods under consideration. Both the Pd/ZnO contacts under consideration are observed to have a good Schottky behaviour under dark condition and a high response to H_2 gas with relatively short response and recovery times. Device fabricated by thermal evaporation shows better performance.

Keywords: Nanocrystalline ZnO thin films, Palladium catalyst, H2 gas sensor, Schottky contacts, Thermal evaporation, Sol-gel

1 Introduction

Hydrogen (H₂) has an important place in modern pollution free fuel technology as an alternative hydrocarbon-free fuel to the fossils fuels. It is a highly combustible and explosive gas with flammability lying in the range \sim (4-47%) by volume¹⁻³ thereby making it more flammable than other commonly used fuels. Thus, the leakage of H₂ in various applications such as transportation and storage of H₂, solid oxide fuel cells, hydrogen engine cars etc. is required to be monitored to avoid a possible explosion. Since H₂ is produced up to (0-2%) due to methane explosion in coal mines⁴, the continuous monitoring and detection of H₂ is an essential requirement in the coal mines to avoid any explosion due to H₂ in the presence of potentially explosive mine atmosphere. Hydrogen detection is also necessary in industrial and domestic environments where clean and pollution free fuels are used. Thus, the development of H₂ gas sensors has become an important area of research in recent times⁴⁻⁸. Among various metal oxides, the ZnO is

considered to be one of the most promising materials for gas sensors especially for H_2 sensing⁶⁻⁸. In general, the nanocrystalline ZnO is preferred over the bulk ZnO because of its higher sensitivity, larger surface area, and better compatibility with other nanodevices than the bulk ZnO. However, the electrical, optical and gas sensing properties of the nanocrystalline ZnO thin films depend on crystal size, surface morphology, crystal orientation, aspect ratio and crystalline density of the nanostructures of the ZnO material. Further, the sensing characteristics of ZnO nanostructures are directly related to its fabrication techniques and operating temperatures $^{8-11}$. In general, small-sized nanoparticles decrease the response time and increase the sensitivity of the ZnO nanostructure based gas detectors¹⁰. The operating temperature plays also a significant role in optimizing the sensitivity of the ZnO thin film based H_2 gas sensors⁶⁻⁸. However, the major limitation of the ZnO thin film based H₂ detectors is the requirement of high operating temperature (normally in the range 200-500°C) for their satisfactory operation⁹. Since H_2 is a highly explosive gas, it may be an important need to develop ZnO thin film based H_2 detectors which can be operated at room temperature.

A number of research papers¹²⁻¹⁶ have already been reported on the fabrication of nanocrystalline ZnO thin films. Among the commonly used deposition techniques including metal organic chemical vapour¹² deposition (MOCVD), pulsed laser deposition¹³ (PLD), thermal evaporation¹⁴, radio frequency sputtering¹⁵ and sol-gel¹⁶, the sol-gel and thermal evaporation methods are more advantageous over the others in terms of low-cost, simplicity in process control and large area deposition. Besides the fabrication technique used for film preparation, the sensitivity, selectivity, responsivity, and recovery time characteristics of the ZnO nanostructure based Schottky contacts can be enhanced by using a noble metals like palladium (Pd) and platinum (Pt) as the Schottky metal¹⁷. The noble metal functions as an electrical contact as well as a catalyst for reducing gas interaction processes in such Schottky contact based gas sensors 5,8,17-19. In the present paper, a comparative study of the room temperature H₂ gas sensing properties of the Pd/nanocrystalline-ZnO based Schottky diodes grown on *n*-type silicon (100) substrates by the thermal evaporation and sol-gel techniques, is reported. Since n-Si/n-ZnO behaves as an ohmic contact²⁰, the use of n-Si in proposed structure can provide the flexibility of integration with present CMOS technology to achieve smart gas sensor. Although, the structure fabricated by thermal deposition technique has already been reported¹⁴, the sol-gel derived ZnO film grown on *n*-Si substrates is believed to be considered first in this paper. Besides gas the sensing properties, the structural, morphological and optical properties of nanocrystalline ZnO thin films grown by the two methods under consideration are also compared in the present paper.

2 Experimental Details

2.1 Preparation of ZnO thin film by thermal evaporation method

The first set of nanocrystalline ZnO thin films used in this study was grown on *n*-Si (100) substrates with a sheet resistivity of 3.6 Ω .cm by using the thermal evaporation method. Ultrapure (~99.99%) ZnO powder (MERK-Chemical Limited, Mumbai, India) was used as the source material. Before deposition, the *n*-silicon substrate was first cleaned in a sequence with acetone, isopropyl alcohol and deionized (DI) water. The DI water (resistivity ~18 M Ω cm) was obtained from the Milli-Q water plant of Millipore, USA. The thermal deposition set-up unit (Model no 12A4D of HINDVAC, India) was used for growing the layer of ZnO thin film. The base pressure of vacuum chamber was maintained at 10⁻⁵ mbar. The substrate was kept at room temperature (27°C) during the deposition of the films. The heating filament used was a conventional molybdenum boat. The thickness of the deposited film was directly monitored by the digital thickness monitor attached to the deposition unit. In order to improve the quality of ZnO thin film and its conductivity, the film was subjected to thermal annealing after deposition at a selected temperature of 450°C in the presence of an Ar environment of a Rapid Thermal Annealing (RTA) chamber¹⁴. The studies on structural and morphological properties of ZnO thin films were carried out after bringing the temperature down to room temperature (27°C).

2.2 Preparation of ZnO thin film by sol-gel method

Besides the nanocrystalline ZnO thin films grown by the thermal evaporation method discussed above, nanostructured ZnO thin films were also grown by spin-coating technique on the same n-Si substrates as used in the thermal evaporation method. Zinc acetate dehydrate $(Zn (CH_3COO)_2 2H_2O)$ (MERCK-Chemical Limited, Mumbai, India) was used as the starting material in our sol-gel method. Isopropyl (MERCK-Chemical Limited. alcohol (C_3H_8O) India) diethanolamine Mumbai, and (HN (CH₂CH₂OH)₂, DEA) (Qualigens Fine Chemicals, Mumbai, India) were used as a solvent and stabilizer, respectively. Zinc acetate dihydrate was first dissolved in a mixture of isopropyl alcohol and DEA solution at room temperature. The molar ratio of DEA to zinc acetate dihydrate was maintained at 1.0 and the concentration of zinc acetate was 0.14 M. The solutions were stirred (magnetically) at room temperature (27°C) for 2 h to yield a clear transparent and homogeneous solution. Then the coating solution (ZnO) was dropped onto *n*-Si substrates which were then rotated at 1000 and 2000 rpm for 15 s for each speed by using a SPM 200 (TSE) spin coater. After depositing one layer of ZnO, the film was pre-heated in air at 250°C for 15 min on a hot plate to evaporate the solvent and remove organic residues. The ZnO film thickness of a layer was estimated from the ellipsometer measurement as ~50-60 nm. The deposition procedures from coating to drying were repeated five times to obtain a ZnO thin film thickness of ~250-300 nm. In order to improve the quality of ZnO thin film and its conductivity, the film was subjected to thermal annealing after deposition at a selected temperature (450°C) in Ar environment of a Rapid Thermal Annealing (RTA) chamber. The deposited ZnO thin film was then left to cool down to room temperature (27°C) before carrying out the structural and optical measurements. The thin films prepared by thermal evaporation and sol-gel methods were labeled as A₁ and A₂, respectively.

2.3 Film characterization

The crystalline orientations of the ZnO thin films were determined by X-ray diffraction (XRD) (Panalytical X'Pert PRO MRD, Netherlands). The surface morphology and elemental composition of the nanocrystalline ZnO thin films were studied by the Scanning Electron Microscopy (SEM) (ZEISS-FESEM) and Energy Dispersive X-ray Spectroscopy (Model:EDX-INCA, Oxford Instruments, UK),respectively. The optical band gaps of the thin films were determined by using the results of photoluminescence (PL) spectroscopy (Perkin Elmer USA made).

2.4 Device fabrication

Figure1 shows the Pd/ZnO/n-Si/Ti/Al device structure considered for the present study. The shadow mask technique was used to deposit Pd (of ~200 nm thickness) with the Schottky contact area $A=7.8\times10^{-3}$ cm² in vacuum coating unit¹⁴. To improve the contact quality and minimizing the interface effect, the fabricated device was annealed in N₂ atmosphere at 450°C for 7 min. A single Pd contact covered a number of closely packed ZnO crystalline particles. The backside of the *n*-type Si (100) substrate was coated with Ti/Al (~200 nm thickness) to realize an ohmic contact. The devices containing, the ZnO thin film grown by the thermal evaporation and sol-gel methods were labeled as A₁₁ and A₂₂, respectively.



Fig. 1 — Device structure

3 Result and Discussion

3.1 Structural and surface morphological properties of ZnO thin films

The structural properties of nanocrystalline ZnO thin films grown by the thermal evaporation and solgel techniques are studied by the XRD analysis as shown in Fig. 2. It is observed from Fig. 2 that both the ZnO thin films are mainly of polycrystalline nature with peaks at (100), (002), (101), (102), (110) (103) and (112). All the diffraction peaks are indexed according to the standard data files^{20,21} (JCPDS file no. 36-1451) which confirm the presence of ZnO thin films with a hexagonal phase. However, the diffraction intensity of the (101) peak was observed to be much higher than that of the (002) peak in the solgel derived film (in A₂ sample) whereas the difference in the intensity between (101) and (002) peaks is very small in case of thermally deposited film (in A₁ sample). In general, the intensities of all peaks in solgel derived films are observed to be higher than their corresponding peaks in the thermally deposited film. The above observation implies that the crystalline quality of the thermally deposited ZnO films is worse than that of the sol-gel derived ZnO films.

The crystallite size (D) can be estimated for both the thermally deposited and sol-gel derived ZnO thin films by the well known Scherer formula²²:

$$D = \frac{c\,\lambda}{\beta\cos\theta} \qquad \dots (1)$$

where β is the full width at half maximum of X-ray peak, λ (=1.54059Å) is the X-ray wavelength and *c* is the correction factor taken as 0.90 in the calculation The structural parameters for both the films extracted from their respective XRD spectra are listed in



Fig. 2 — XRD spectrum of ZnO thin fabricated by sol-gel and thermal evaporation method

Table 1. From Table1, it is evident that the crystal size of the sol-gel derived ZnO films in A_2 is larger than that of the thermally deposited films in A_1 sample. Clearly, the thermally evaporated film has larger surface area than that of the films prepared by using the sol-gel technique.

Further, the higher values of the full width at half maximum (FWHM) of the prominent peaks of sample A_1 (Table 1) indicate a poorer crystalline quality of the thermally deposited ZnO film in A_1 samples as compared to that of the A_2 samples²².

The SEM image and EDX spectrum of the nanostrctured ZnO thin films grown by the thermal

| Table 1 — Crystalline data for ZnO thin film fabricated by the | | | | | | |
|----------------------------------------------------------------|-------|----------------|--------|----------|--|--|
| sol-gel and thermal evaporation method | | | | | | |
| Sampla | Daala | Deals Desition | EWIIM | D(nm) | | |
| Sample | геак | | | D (IIII) | | |
| | | (deg) | (rad) | | | |
| ٨ | 100 | 31 37 | 0.0088 | 16.4 | | |
| A 11 | 100 | 51.57 | 0.0000 | 10.4 | | |
| | 002 | 34.42 | 0.0105 | 13.8 | | |
| | 101 | 36.26 | 0.0097 | 15.0 | | |
| A ₂₂ | 100 | 31.77 | 0.0066 | 21.0 | | |
| | 002 | 34.42 | 0.0094 | 15.4 | | |
| | 101 | 36.23 | 0.0076 | 19.2 | | |
| | | | | | | |

evaporation method are shown in Fig. 3(a and b) whereas those of the films grown by the sol-gel technique are shown in Fig. 3(c and d), respectively. The SEM images of Fig. 3(a and c) show that the thermal evaporated film has a smaller particle size than that of the films grown by the sol-gel method. Note that the results are in consistence with the crystallite sizes estimated from the XRD analysis as discussed earlier. This clearly shows that the thermally deposited ZnO films have higher surface-to-volume ratio thereby having better gas sensing properties¹ than that of the sol-gel derived films.

The EDX data shown in Fig. 3(b and d) show that the peak corresponding to 0.52 keV represents the presence of oxygen (O) whereas other two peaks at 1.012 and 8.63 keV represent the Zn element in the films grown by the two methods under consideration. Note that the peak at around 1.486 keV corresponds to Si substrates on which the films are fabricated. Clearly, the EDX results also confirm that the thinfilms grown by both the thermal evaporation and solgel methods contain the Zn and O elements of the ZnO nanostructures under consideration.



Fig. 3 — (a, b) SEM and EDX of ZnO thin films fabricated by thermal evaporation; (c, d) SEM and EDX of ZnO thin films fabricated by sol-gel method

3.2 Optoelectronics properties of ZnO thin film

The room temperature PL spectra obtained by using the He-Cd (Perkin Elmer USA made) laser of 325 nm emission line to study the optical properties of nanocrystalline ZnO thin films under consideration are shown in Fig. 4. The UV emission is mainly due to the excitonic transition between valence and conduction band whereas the broad peak is believed to be due to the defects states in ZnO films^{22,24}.

The PL spectrum of Al sample shown in Fig. 4 shows a broad ultraviolet (UV) emission peak at a wavelength of 380 nm and some defect related emission peaks in the 450-550 nm range (i.e. yellow-green emission) of the visible region. The UV emission is mainly due to the excitonic transition between valence and conduction band whereas the broad peak is believed to be due to the defects states in ZnO films^{22,24}. There are certain intrinsic defects presents in undoped ZnO as Zn vacancies, interstitial oxygen and antisite oxygen²⁴.

The electrical and optical properties of ZnO thin film are greatly influenced by these intrinsic defects which may be either from the acceptor or donor levels^{25,26}. The defects related to the oxygen vacancy may show a green or yellow emission in the PL spectrum¹³. On the other hand, the PL spectrum of A_2 sample shows one strong photoluminescence emission peak and some weak peaks in the visible region. The strong peak of ZnO is localized at around 378 nm which corresponds to the band-to-band transition. The other peaks in the visible region belonging to the yellow-green emission are due to defects in the solgel derived ZnO film. However, higher intensities of the defect-related peaks in the visible region of the A_1 sample imply that more crystal defects are present in



Fig. 4 — Photoluminance of ZnO thin film by sol-gel and thermal evaporation method

the thermal derived films in A_1 samples than the solgel derived ZnO films. The weak intensity of the near-band-edge and broadening of the visible band are attributed to the weak polycrystalline nature of the films (in A_1 samples). The optical band gaps of the two types of ZnO thin films estimated from the PL spectra are 3.26 eV and 3.28 eV for A_1 and A_2 samples, respectively which are close to the value reported by others for bulk²⁵ ZnO. This indicates that the optical property of ZnO thin film is dependent on the deposition techniques.

3.3 Gas sensing of Pd/ZnO interface

Homemade gas sensing systems interfaced with semiconductor parameter analyzer (Agilent, USA makes model no. 1500A) were used to perform the electrical and H_2 gas sensing characteristics of fabricated Schottky devices at room temperature. The I-V characteristics of both kinds of nanostructured-ZnO based Schottky diodes fabricated by the two methods under consideration were carried out for 0 and 1% H_2 gas concentrations by varying the *dc* bias voltage in the range of -1 V to 2 V as shown in Fig. 5. The results in Fig. 5 clearly show the typical



Fig. 5 — Characteristics of the device A_{11} and A_{22} in 0% H_2 and 1% H_2 at room temperature (a) I-V and (b) ln(J)-V

rectifying behaviour in the both types of Pd/ZnO Schottky diodes. No breakdown is observed in the reverse bias operation in the 0 V to -1 V bias voltage range for both sets of devices A_{11} and A_{22} . Further, the forward bias current is observed to be increased with the increase in hydrogen gas concentration from 0 to 1% in the gas sensing chamber for both the devices. The above observations clearly indicate that both the devices A_{11} and A_{12} can be used to detect hydrogen gas.

The hydrogen gas sensing mechanism of Pd/ZnO contact can be explained by basic properties of the metal semiconductor contact as follows: The larger work function of Pd than that of the ZnO forms a potential barrier at the Pd/ZnO interface²⁷. This difference in work function also generates a depletion region in the ZnO side due to the carrier transport from the semiconductor to the metal side under zero bias condition. Note that ZnO is inherently an *n*-type semiconductor and hence the depletion region is increased (decreased) with the increase (decrease) in the -ve (reverse bias) potential at the Pd contact electrode with respect to the ZnO. Thus, in the absence of H₂ gas, the I-V characteristics can be described by the commonly used thermionic emission process at the Pd/ZnO Schottky junction²⁸ and the forward bias current is increased with applied forward bias (+ve) voltage of the Pd with respect to ZnO due to the reduction in the depletion region described above. However, in the presence of H₂, the gas is diffused into Pd thin film surface and hydrogen molecules are dissociated into hydrogen atoms. Some hydrogen atoms react with the thin Pd metal layer to form the palladium hydride near the metalsemiconductor interface which lowers the work function of Pd metal at the Pd/ZnO junction thereby decreasing the barrier height of the Pd/ZnO Schottky contacts^{7,8,28}. Clearly, the decrease in the barrier height increases the forward bias current²⁸ as shown in Fig. 5.

To get the insight of sensing mechanism of the Schottky diodes under consideration, the diode parameters like ideality factor, saturation current and barrier height are estimated for 0% and 1% H₂ gas concentrations. Note that the *I-V* relation of the *n*-ZnO/metal Schottky diode can be described by using the thermionic emission model^{17,27-29} as :

$$I = \left[AA^*T^2 \exp\left(-\frac{q\phi_b}{kT}\right)\right] \left[\exp\left(\frac{qV}{\eta kT}\right) - 1\right] \qquad \dots (2)$$

where η is the ideality factor, *k* is the Boltzmann constant, *T* the absolute temperature, *A* the rectifying contact area, *A*^{*} is the effective Richardson constant²⁸ (theoretically *A**=32 A cm⁻²K⁻² for *n*-ZnO) and ϕ_b is the barrier height. Assuming $qV/\eta kT>>1$, Eq. (2) can be approximated as:

$$I = I_s \exp\left(\frac{qV}{\eta KT}\right) \qquad \dots (3)$$

where I_S is saturation current given by:

$$I_{S} = \left[AA^{*}T^{2} \exp\left(-\frac{q\phi_{b}}{kT}\right) \right] \qquad \dots (4)$$

Following the similar methodology^{28,29}, the diode parameters for both the devices are extracted from their respective characteristics as listed in Table 2.

It is clearly observed from Table 2 that the barrier height of both the devices is found to be decreased with the increase in the H_2 concentration from 0% to 1%. The barrier lowering due to the palladium hydride layer formation at interface is responsible for increase in forward current of both devices A_{11} and A_{22} as discussed earlier. The extracted values are found to be in fairly good agreement with those reported by other researchers²⁸ for both devices A_{11} and A_{22} .

However, it may be noted that the measured barrier heights are smaller than the theoretical value of 1.02 eV computed from the Schottky–Mott $model^{29}$. The observation is in coherence with the observation reported by other researchers^{27,28}. This implies that the thermionic emission theory is inefficient to explain accurately the transport mechanism in the Pd/ZnO based Schottky junction diodes. There are several possible explanations including the image force, interfacial layer, tunneling through Schottky barrier and generation/recombination of electron-hole pairs in the depletion region are some main effects which make the barrier height as a function of applied bias and hence play important roles in lowering the barrier height of the Pd/ZnO Schottky contacts. The lowering in barrier height due to hydrogen gas enhances the Table 2 — Different parameter of two devices A₁₁ and A₂₂ for 0%

| and 1% hydrogen | | | | | | |
|-----------------|----------------|------------------------------------------|-----------------------|------------------------|--|--|
| Device | H ₂ | Ideality factor (A/cm ²) | Saturation current | Barrier height (eV) | | |
| A ₁₁ | 0% | 4.5 | 1.06×10 ⁻⁶ | 0.59 | | |
| | 1% | 4.6 | 4.54×10^{-6} | 0.58 | | |
| A ₂₂ | 0% | 4.3 | 1.04×10^{-6} | 0.68 | | |
| | 1% | 3.3 | 3.14×10^{-5} | 0.65 | | |



Fig. 6 — Response versus biased voltage graph for the two devices A_{11} and A_{22}

reverse saturation current as shown in Table2. The measured ideality factors are much larger than unity in both the devices due to the possibility of barrier inhomogeneity²⁷ in the Pd/ZnO Schottky contacts. However, the differences in the estimated values of the barrier height, saturation current and ideality factor between A₁₁ and A₂₂ may be attributed to the differences in the surface morphology and crystalline properties of the ZnO thin films due to two different fabrication processes namely the thermal evaporation and sol-gel methods used in A₁₁ and A₂₂, respectively. The relative response (sensitivity) in case of the Schottky diode can be defined^{8,17} as :

$$S = \frac{I_g - I_a}{I_a} \qquad \dots (5)$$

where I_a is the forward current in air (with 0% H₂) while I_g is the forward bias current in the presence of H₂ gas for a fixed bias condition of the device.

Figure 6 shows the maximum relative response for 1% H₂ concentration at room temperature for A_{11} and A₂₂ samples. The biased voltages of 0.7 V and 0.4 V are used as the operating voltages of A_{11} and A_{22} samples, respectively since the maximum relative responses of A₁₁ and A₂₂ are achieved at the respective bias voltages. It is observed from Fig. 6 that the response of A₁₁ at room temperature is approximately twice of that of A_{22} . The larger response in A_{11} as compared to A_{22} is attributed to the poorer crystal quality, smaller grain size, and smaller particle size¹⁰ of the ZnO film grown by the thermal evaporation method as evident from the XRD and SEM results discussed earlier. The above observed response of the Pd/ZnO Schottky diode fabricated by the thermal deposition method (i.e. A₁₁) under consideration is



Fig. 7 — Response versus time graph for the device A_{11} and A_{22}

more than twice of that of the Pd-Ag/ZnO/PS (porous Si)/p-Si/Al device reported for hydrogen detection at room temperature⁸. Note that A₁₁ is a much simpler device structure than the Pd-Ag/ZnO/PS (porous⁸ Si)/p-Si/Al for hydrogen detection at room temperature.

Finally, the transient responses of the A_{11} and A_{22} devices are compared in Fig. 7 for 1% hydrogen in air at room temperature. The response recovery time is defined as the time corresponding to the 67% of the saturation value⁸. It is observed that A_{11} possesses superior recovery time characteristics than that of the A_{22} . The improved response and recovery of the device A_{11} are mostly dependent on the method of fabrication of ZnO thin films, surface to volume ratio and crystal quality of the films.

4 Conclusions

Thermal evaporation and sol-gel techniques have been used for the fabrication of nanocrystalline-ZnO thin film based Pd/ZnO Schottky diodes on n-Si substrate for hydrogen gas detection. The structural and optical properties of ZnO thin films fabricated by both techniques have been investigated systematically by XRD, SEM, EDX and PL spectra. The results show that the thermally evaporated thin films have smaller grain size (i.e. larger surface-to-volume ratio) with lesser polycrystalline nature than those of sol-gel derived films. It is also found that the fabrication methods significantly affect the structural and optical properties of the obtained nanocrystalline ZnO thin films. The *I-V* and hydrogen gas sensing measurements reveal that both the devices can be used for the detection of hydrogen gas at room temperature. However, the hydrogen gas sensing response of Pd/ZnO thin films based Schottky diode fabricated by the thermal evaporation is higher than that of the sol-gel derived films. The study could be very much useful for the designing of Pd/nanocrystalline-ZnO Schottky diode based hydrogen gas detectors operating at room temperature.

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