

Response letter

Dear Reviewers

First of all, we would like to express our great thanks to you for your carefully reading our manuscript and providing us very helpful suggestions. We appreciate your comments very much.

According to your requirements and suggestions, we have carefully answered the questions mentioned by reviewers, and revised the manuscript in red color.

We would like to resubmit the revised manuscript to *Materials*. Thank you for your consideration and time.

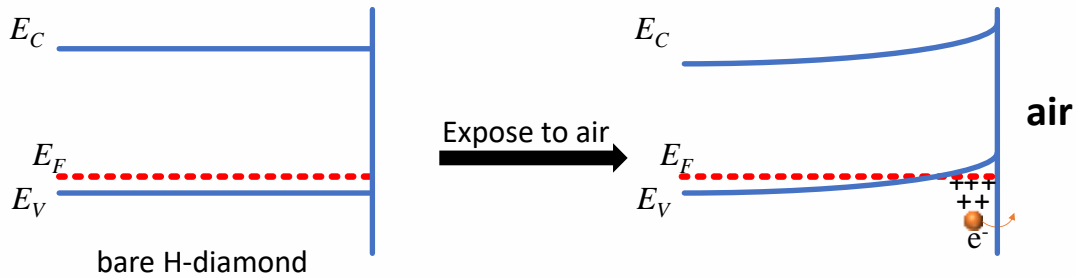
Best regards,

Genqiang Chen

Reviewer 1

1. Since this is a normally-on device, it is assumed that there is a conducting channel under the gate. Explain how this hole-channel is formed? A simple explanation would be enough.

Thanks for your suggestion. When a hydrogen-terminated diamond is exposed to air, some negatively charged adsorbates form at diamond surface, such as OH^- , HCO_3^- and CO_3^{2-} etc. Electrons at diamond surface can transfer to these adsorbates, thus the compensating hole-channel beneath diamond surface is formed. The energy band diagram is shown as follows,



In addition, Ref.4 was cited to interpret how the hole channel is formed. And, the sentence, page 1 line 34, “Thanks to the hydrogen-terminated diamond (H-diamond) which can form the two-dimensional hole gas (2DHG) after hydrogen plasma treatment beneath the surface” was revised to be “*Thanks to the hydrogen-terminated diamond (H-diamond) which can form the two-dimensional hole gas (2DHG) beneath the surface when it is exposed to air*”, which was used red font in the revised manuscript

4 F. Maier, M. Riedel, B. Mantel, J. Ristein, and L. Ley, *Physi. Rev. Lett.* 85, 3472 (2000).

2. Since this is an interface device, it would help the reader to see an energy band diagram for the device. Is there a quantum well at the interface due to surface band bending? How does the band look?

We appreciate your precious advice. Since the energy band energy diagram of Normally-On ALD- Al_2O_3 /H-diamond is well-known, we drew the energy band diagram of ALD- Al_2O_3 /H-diamond here as follows,

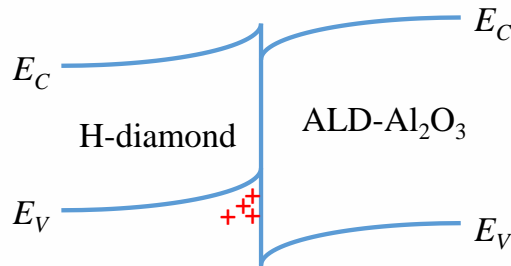


Fig2 (b) energy band diagram of H-diamond/ Al_2O_3 without gate bias

Due to surface band bending, there is a quantum well at diamond surface. We revised the Fig.1 and Fig.2 in the revised manuscript. The energy band diagram is shown in Fig.2 (b). And, a simple illustration “Fig.2 (a) shows the schematic diagram of the MOSFET. The gate width (WG), gate length (LG) and distance source/drain are 100, 2, and 20 μm , respectively. L_{SG} and L_{GD} are 9 μm . Fig.2(b) illustrates energy band diagram of H-diamond/ Al_2O_3 . The 2DHG under diamond surface is accumulated due to negatively charged adsorbates, which leads to the energy band upward bending at diamond surface” for Fig.2 (b) was added into page7, line 100~104.

3. The dielectric is grown using ALD, which is assumed to grow materials with low defect densities. Why are there $1.66 \times 10^{13}/\text{cm}^2$ and trapped charges of $4.03 \times 10^{12}/\text{cm}^3$ these are very high values for the device to be stable? Can you explain?

Thanks for your professional comments .In fact, defect densities in ALD-Al₂O₃ strongly depends on synthesis temperature. In this work, the dielectric deposition began at 90°C. This temperature is relatively low so that there are lots of defects in Al₂O₃. Thereby, a high density of trapped charges and interface states density exists. According to literature¹, trapped charges can induce the threshold voltage instability after a high gate bias sweeping. In our experiment, the interval between two measurement was 10 h minimize this affect, and a little difference of $V_{th}(<0.2\text{ V})$ was observed.

¹ Z. Chen, X. Yu, J. Zhou, S. Mao, Y. Fu, B. Yan, R. Xu, Y. Kong, T. Chen, Y. Li, and Y. Xu, Appl. Phys. Lett. **117**, (2020).

² Y. Li, J.F. Zhang, G.P. Liu, Z.Y. Ren, J.C. Zhang, and Y. Hao, Phys. Status Solidi - Rapid Res. Lett. **12**, 1 (2018).

³ Y. Sasama, T. Kageura, M. Imura, K. Watanabe, T. Taniguchi, T. Uchihashi, and Y. Takahide, 1 (2021).

4.Any evidence for the validity of the explanation for the decrease of the negative charges with annealing?

Thanks for your suggestion.

The decrease of negative charges can be explained in two ways.

1. It is well-known that a 2D hole channel formed at H-diamond surface is induced by surface adsorbates. These adsorbates commonly are present negative polarity, such as OH⁻, HCO₃⁻ and CO₃²⁻ etc. It is negative charges that induce holes beneath H-diamond surface. According to the *negatively shifted threshold voltage* after annealing, it is confirmed that the hole density decreased. Thus, we could conclude that the negative charges decreased.
2. In another way, the decreased negative charges with annealing can be confirmed by the negatively shifted flat band voltage. In accordance with equation.2, the decreased V_{FB} results in the decreased quantities negative charges.

5.Any evidence for the validity of the explanation for the increase of mobility and the subsequent decrease with annealing?

Thanks for your comments. In order to make sure the mobility was worked out with **a same carrier density**, we used equation (3) to calculate the mobility **at $V_{th}-V_{GS} = 2\text{ V}$** .

$$I_{DS} = \frac{W_G \mu_{eff} C_{ox} (V_{GS} - V_{th})^2}{2L_G} \quad (3)$$

The current density I_{DS} at $V_{th}-V_{GS} = 2\text{ V}$ is 8.6 mA/mm. After 150 °C annealing process, the current density at $V_{th}-V_{GS} = 2\text{ V}$ is 10.8 mA/mm, and it is 5.9 mA/mm after 200 °C annealing process. According to ohm's law and definition of mobility, the current density can be given by

$$\mathbf{J} = e\rho\mu$$

Where the J is current density , the p is carrier density ,μ is mobility, and e is electron charge. Thereby, the increase of current density and subsequent decrease with annealing could be the evidence for our explanation.