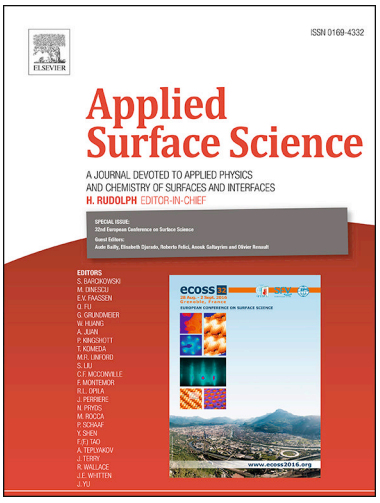

First-principles study of oxygen-related defects on 4H-SiC surface: The effects of surface amorphous structure

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PII: S0169-4332(18)32488-7
DOI: https://doi.org/10.1016/j.apsusc.2018.09.072
Reference: APSUSC 40376

To appear in: *Applied Surface Science*

Received Date: 2 August 2018
Accepted Date: 8 September 2018

Please cite this article as: Y-i. Matsushita, Y. Furukawa, Y. Hijikata, T. Ohshima, First-principles study of oxygen-related defects on 4H-SiC surface: The effects of surface amorphous structure, *Applied Surface Science* (2018), doi: https://doi.org/10.1016/j.apsusc.2018.09.072

First-principles study of oxygen-related defects on 4H-SiC surface: The effects of surface amorphous structure

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(Dated: September 10, 2018)

We report our first-principles calculations that clarify the electronic states of oxygen-related defects in the 4H-SiC bulk and on the 4H-SiC surface and how they are affected by the surface amorphous structure due to oxidation. It is experimentally reported that thermally oxidized 4H-SiC contains an abundant amount of single-photon sources on its surface (surface SPSs) and that their emitting wavelengths have variance. However, the microscopic mechanism is not clarified yet. In our work, we demonstrate that the energy levels of the oxygen-related defects on the surface are altered sensitively by the local atomic structure of the amorphous surface leading to variations in the wavelengths.

Single-photon sources (SPSs) are an essential element for quantum information processing and quantum computing technology. When an electron pumped up to an excited state by light goes back to the ground state, the SPS generates a single photon with a wavelength corresponding to the HOMO(highest-occupied-molecular orbital)-LUMO(lowest-unoccupied-molecular orbital) gap. For practical application of SPS devices, solid devices are desirable because of its stability and mobility. Some defect structures in many semiconductor materials are known to date to work as SPSs such as NV^- center in diamond and Zn vacancy (V_{Zn}) in ZnO crystal[1]. Recently, SiC is getting much attention for SPS devices, since SiC contains various types of intrinsic defects that behave as an SPS. Many of SPSs in SiC bulk are already identified, such as silicon vacancy (V_{Si})[2,3], carbon-silicon di-vacancy ($V_C V_{Si}$)[4], and antisite-vacancy pair ($C_{Si}V_C$)[5]. In addition, it is reported that the brightness of SPSs in SiC can be controlled electrically by embedding the SPSs in SiC-MOS (metal-oxide semiconductor) devices. This opens a possibility of new optoelectronic devices[6–9].

Interestingly, a large number of bright SPSs have been reported to emerge on the surface (surface SPSs) of SiC through thermal oxidation on Si-[7,10,11] and C-face[8,10] both. These SPSs have different characters from other known SPSs. One interesting point is that the surface SPSs are more than twice as bright as even NV^- center in diamond[7]. Another striking feature is that there is a certain amount of variance in their emitting wavelengths of the zero-phonon line (ZPL)[8,11]. However, the microscopic mechanisms of the variance in the ZPL are not clarified yet. One possible scenario is an interplay between an SPS and a stacking fault[7]: The wave function of the excited state around a surface SPS is modulated by the stacking fault depending on the SPS position in real space. The stacking fault structure in SiC surface is reported both experimentally[12,13] and theoretically[14] and such interaction between a surface SPS and a stacking fault is likely. However, it is reported that there is no clear relation observed between the SPS position near the single Schottky-stacking fault (SSSF) and its ZPL[15]. Only brightness is affected by the SSSF. This fact might show that the wide variety of ZPL of surface SPSs cannot be explained by the stacking fault alone. Microscopic understanding of the variances in the ZPL of the surface SPSs is yet to be achieved.

In this work, we propose another possible mechanism to explain the variances in the ZPL of surface SPSs on the basis of the density-functional theory (DFT)[16] with the Kohn-Sham scheme[17]. The surface of a thermally oxidized SiC is thought to be amorphous through chemical absorption of O atoms, forming complex atomistic structures. Therefore, the different local atomic structures near a surface SPS can cause changes in the ZPL of the surface SPS in the similar way to a defect level in amorphous SiO_2[18,19]. The thermal process is conceivable to introduce many O-related defects (O-defects) on the surface having a possibility of behaving as SPSs. Furthermore, a recent experiment done by us points out that the surface SPSs incorporate O atoms by using the oxygen isotope[20]. Therefore, we focus on O-defects on the surface of 4H-SiC in this work. Also, we have investigated the effects of distortion on the variances of the ZPL. It is because the distortion in SiC/SiO_2 interfaces is observed in experiments[21,22] and is also a conceivable factor to affect the ZPL of the surface SPSs.

All calculations, including both the structural optimizations and the static electronic-structure calculations, have been performed by Vienna *ab-initio* Simulation Package (VASP) with PAW method[23,24] using PBE exchange-correlation functional[25] in the generalized gradient approximation (GGA). Note that while the GGA underestimates band gaps by about 50%, the amount of energy-level shifts can be evaluated quantitatively. In fact, thought the calculated band gap of 4H-SiC is underestimated to 2.4 eV in comparison to experimental value 3.3 eV, the calculated band-gap variations among SiC polytypes are well reproduced[26]. We adopted a slab model with 5 bilayer thickness to simulate the amorphous