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Comparison of the Stability of 2H Nanosurfaces by the Adsorption of Small Molecules : A DFT Study

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ABSTRACT

Density functional theory (DFT) calculations were carried out to understand the structural stability of 2D nanosheets of gold and silver in hexagonal phase of 2H by the adsorption of small molecules. In this work, we have obtained the bonding and adsorption properties of such small molecules as H_2O, H_2O_2, and C_2H_5OH on 2H phase of gold and silver nanosurfaces, through DFT method using Quantum Expresso (QE) code. The high absorption energy values of (-2.45 eV, -2,46 eV, -2, 41 eV) for H_2O, H_2O_2, and C_2H_5OH molecules on 2H-Au surfaces, respectively obtained than that of 2H-silver surfaces that the interaction between small molecules and both 2H nanosurfaces corresponds to physisorption. However, during the adsorption, the gold surface in the 2H phase (2H-Au) seems to preserve its atomic structure, while 2H-Ag surface changes from 2H to the fcc structure. Based on the analysis of electronic and physicochemical properties, the composite systems of 2H-gold/2H-silver-small molecules exhibit semiconductor behaviour. While 2H-Ag surfaces have short recovery time values for hydrogen peroxide (H_2O_2), this time is quite long for 2H-Au surfaces. Because of the long recovery time, Au-2H reported surfaces can be a candidate for possible applications of viral capture. Thus, the reported results are significant, and they would stimulate the experimental and further studies.

Keywords: 2H-Gold, 2H-Silver, nanosheets, Water, Ethanol, Hydrogen Peroxide, Anti-Viral 2D-Materials

I. INTRODUCTION

Recently, the great progress has been made in biomedical, biological, and chemical sciences; etc., owing to their size dependent properties [1], structural and electronic properties [2,3,4]. Also, a specific group of two-dimensional (2D) nanomaterials has drawn attention for physicochemical properties and extensive different applications [5,6,7]. Despite the increasing interest, there are still unknown areas.

Due to its tunable physical and chemical properties humanity has interested in gold and silver for ages. Also, especially noble metals in nanomaterials [8,9] like gold (Au) and silver (Ag) the size affects the physical, chemical and mechanical properties of materials. Not only as enhancing the activity of drugs because of its antimicrobial properties but also surface plasmon resonance property makes it top materials as biosensor [1,10]. Additionally, the silver has biomedical potentials such as drug delivery, medical imaging, antibacterial agent [11,12] and molecular diagnostics [13]. Gold and silver nanoparticles have an antiviral activity against many types of bacteria [14] and fungi [15], and intracellular gene regulation [16].

Gold nanomaterials generally adopt thermodynamically stable face-centered cubic (fcc) phase [17,18]. Silver clusters smaller than 5 nm adopt icosehedral structure, while the clusters larger than 9 nm adopt face-centered cubic structure [19]. In its bulk form, the equilibrium crystal structure for silver is fcc but in nanosize other polytypes of silver have been observed [20].

2H phase nanomaterials are yet to be realized, so in this paper we study the behaviour of one of gold phase Au-2H [17] and one of the polytype of the silver Ag-2H [19] to explore their intriguing properties, the adsorption properties of small molecules on 2D nanosheet and other physicochemical properties with aim of the potential applications.

II. METHODS AND MATERIAL

All calculations we employed were performed using the Quantum-ESPRESSO suite of electronic structure codes, version 5.2.1 [21]. The Perdew-Burke-Ernzerhof generalized gradient approximation (GGA-PBE) [22] has been used for the exchange-correlation, and to deal with electron-ion core interactions the projected augmented wave method (PAW) has been employed; namely, Au.pbe-n-kjpaw_psl.1.0.0.UPF for gold, Ag.pbe-n-kjpaw_psl.1.0.0.UPF for silver, C.pbe-n-kjpaw_psl.1.0.0.UPF for carbon, O.pbe-n-kjpaw_psl.1.0.0.UPF for oxygen and H.pbe-kjpaw.UPF for hydrogen. These pseudopotentials are available from the Quantum ESPRESSO web site [21]. A cutoff energy of 45 Ry was selected for the plane waves used to expand the Kohn-Sham orbitals, and 350 Ry for the charge density. The Brillouin-zone was sampled with a 3 × 3 × 1 Monkhorst-Pack grid. van der Waals corrections has been taken into account using the Grimme-D3 method [21].

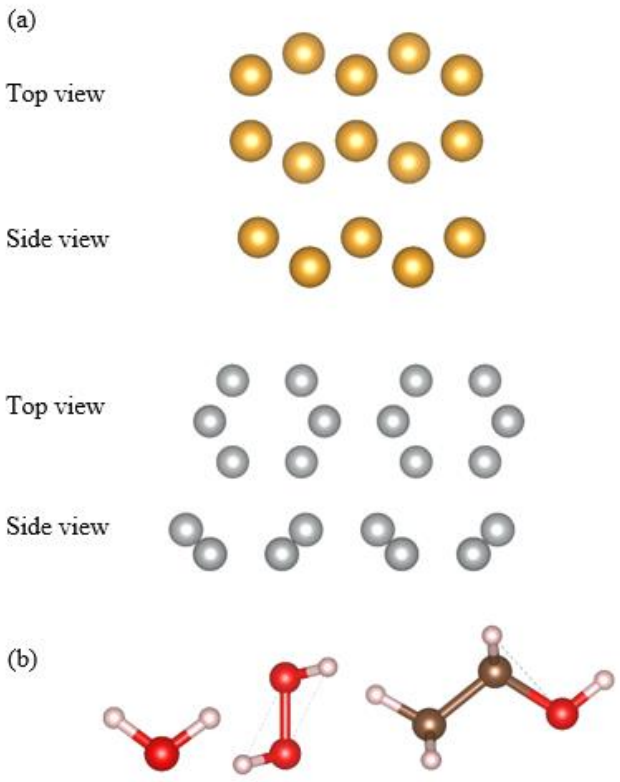


Figure 1: Optimized structures of (a) Au_{2H} and Ag_{2H} (top and side view) and (b) H_2O, H_2O_2, and C_2H_5OH molecules, respectively. Gold, silver, carbon, oxygen and hydrogen atoms are represented by yellow, gray, brown, red and pink spheres, respectively.

Firstly, 2H phase of gold and silver nanosheets are created with the VESTA programme and isolated Au-2H and Ag-2H nanosheets and the other small molecules, H_2O, H_2O_2, C_5H_2OH, are optimized using Quantum-ESPRESSO. Then the small molecules are

placed the preferred position on the Au-2H and Ag-2H nanosheets. While the structure of Au-2H has 10 atoms, the structure of Ag-2H nanosheet contains 12 atoms. 2D Au-2H and 2D Ag-2H nanosheets, and small molecules, H_2O, H_2O_2, C_5H_2OH, are shown in Fig. 1.

Adsorption energies were computed as follows:

$$E_{ads} = E(complex) - E(nanosheet) - E(molecule)$$

where E(nanosheet), E(molecule) and E(complex) are the total energies of nanosheet, molecule and complex system including nanosheet with the molecule. Before the optimization performed, the structures are shown in Fig. 2.

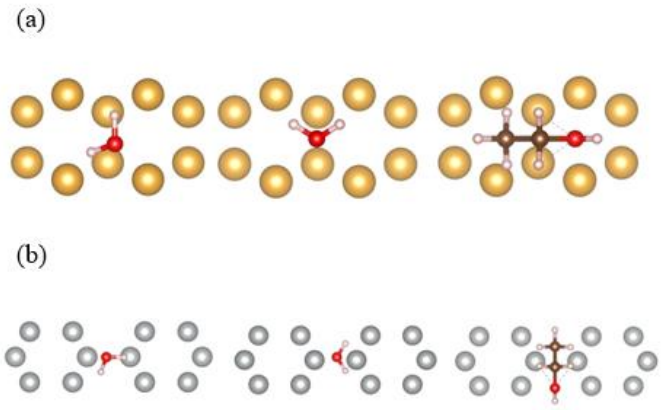


Figure 2: Top views of structures for H_2O, H_2O_2, and C_2H_5OH molecules on (a) Au_{2H} and (b) Ag_{2H} nanosheet before the optimization, respectively.

The electrical conductivity and sensitivity of a material can be determined as:

$$\sigma \alpha \exp(-E_g/2k_bT)$$

where E_g is the bandgap, T is the temperature, k_b is the Boltzmann constant and the sensitivity of a material as:

$$\%S = (|\sigma_{nanosheet} - \sigma_{complex}| / \sigma_{nanosheet}) * 100$$

where $\sigma_{nanosheet}$ is the conductivity of nanosheet, $\sigma_{complex}$ is the conductivity of complex system.

Band gap energy and the percentage change of energy gap can be expressed as:

$$E_g = E_L - E_H$$

$$\%\Delta E_g = 100 * (E_{g\,(complex)} - E_{g\,(nanosheet)}) / E_{g\,(nanosheet)}$$

where E_L is lowest unoccupied molecular orbital (LUMO), E_H is highest occupied molecular orbital (HOMO) and $\%\Delta E_g$ is the percentage change of energy gap E_g of complex after adsorption with respect to the bare nanosheet.

The main purpose of the density functional theory of chemical reactivity is that links of the chemical potential of DFT with the first derivative of the energy with respect to the number of electrons. Hence the negative of the electronegativity is the remark established by Parr, Donnelly, Levy and Palke, the negative of the electronegativity (χ) [23,24];

$$\mu = (\partial E/\partial N)_V = -\chi$$

The parameters such as electronegativity (χ), global hardness (η), chemical potential (μ), global electrophilic index (ω), spherical softness (S), nucleofugality ΔE_n and electrofugality ΔE_e and electronic charge (ΔN_{max}) are related to HOMO and LUMO energies.

The recovery time (τ) can be described as follows:

$$\tau = \upsilon^{-1} \exp(-E_{ads}/kT)$$

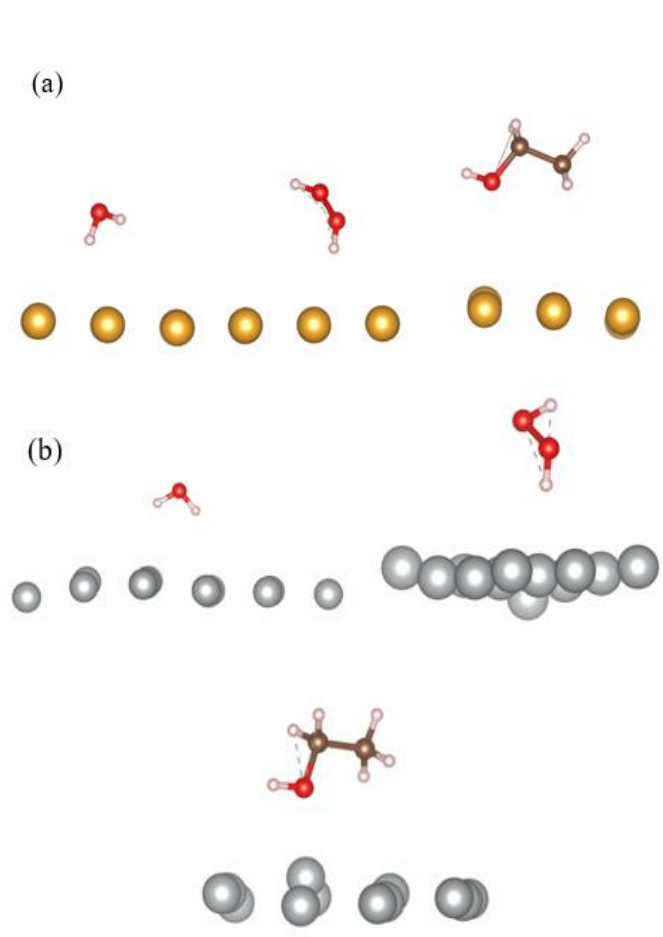
where υ is the attempt frequency, k is the Boltzmann constant and T is temperature. Under UV vacuum light ($\upsilon \sim 3x\ 10^{12}\ sn^{-1}$) at 298K and UV radiation ($\upsilon \sim 10^{16}\ sn^{-1}$) at 398K is applied to recover of the molecule from the studied surfaces [25].

III.RESULTS AND DISCUSSION

Not only surface disinfection properties but also its anti-viral properties like hydrogen peroxide, ethanol is used in hospitals and medical practices. Furthermore, there are many articles that have been studied about the water's antibacterial and antiviral effectiveness. Therefore, we wanted to compare and discuss the antiviral features of these molecules and focused on the adsorption of them on nanosheet.

After the optimization, the zigzag structure of the 2D nanosheet are not preserved and turned into more stable phase in all complexes represented in Fig. 3.

The optimized geometries of the complexes and the distances of gold and silver atoms on the nanosheets in all complex structures are shown in Fig. 4. After the interaction with H_2O, H_2O_2, and C_2H_5OH molecules, the nanosheets turned into stable phase face-centered cubic structure and the distances for all complex structures change between 2.62-2.87 Å. The lattice constants for gold and silver are 4.0796 Å and 4.0855 Å in Inorganic Crystal Structure Database (ICSD). If the distances between atoms are calculated for fcc structure of gold and silver by ICSD data, the distances are 2.8847 Å and 2.8888 Å for gold and silver, respectively.



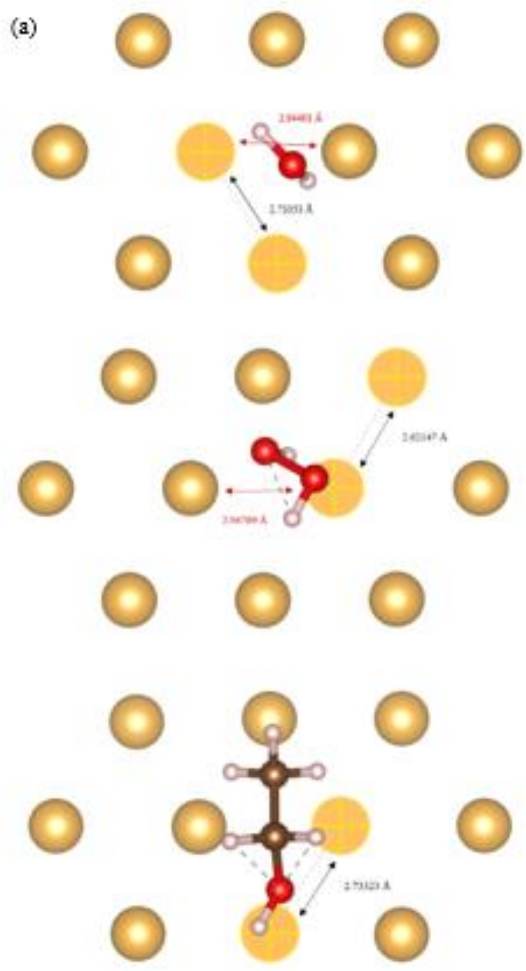


Figure 3: The examples of changing structure of (a) Au_{2H} and (b) Ag_{2H} nanosheets after the interaction with H_2O, H_2O_2, and C_2H_5OH, respectively.

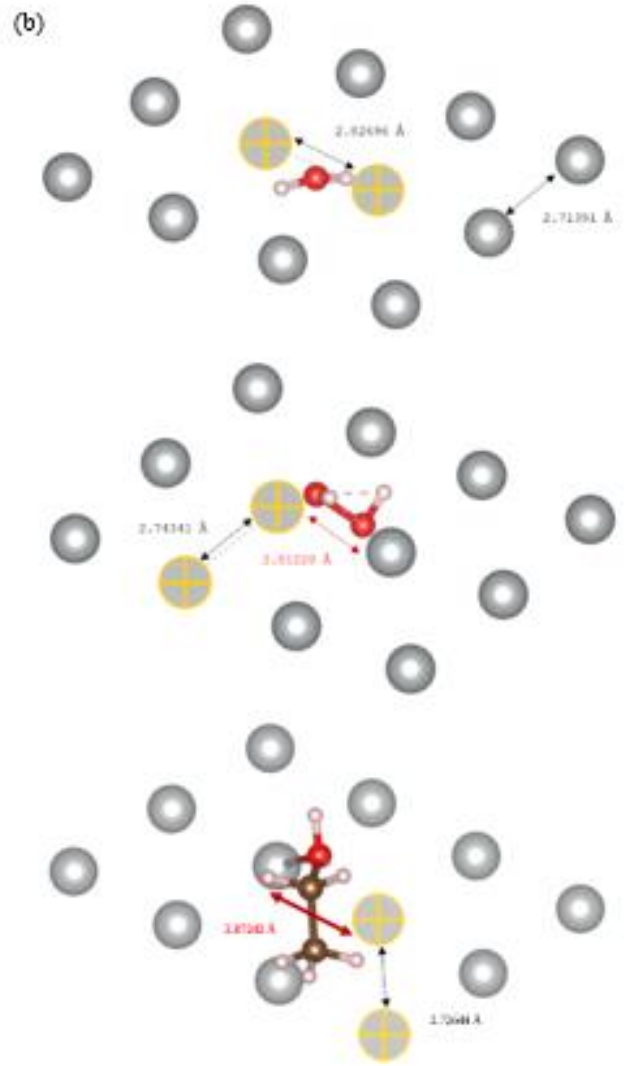


Figure 4: After the optimization the distances between (a) gold atoms on Au2H nanosheet and (b) silver atoms on Ag2H nanosheet

After the optimization the calculated adsorption energy values in eV are shown in Tab. 1. 2D Au-2H nanosheet adsorbs molecules more than 2D-Ag-2H nanosheet. Negative adsorption energy suggests that the adsorption process is exothermic.

Table 1 The calculated adsorption energy values (in eV) of H_2O, H_2O_2, C_2H_5OH on 2D Au_{2H} and Ag_{2H} nanosheets.

Structure	Energy values (eV)
$E_{ads}(H_2O)_{Au\text{-}2H}$	-2.44635
$E_{ads}(H_2O_2)_{Au\text{-}2H}$	-2.46071
$E_{ads}(C_2H_5OH)_{Au\text{-}2H}$	-2.40893
$E_{ads}(H_2O)_{Ag\text{-}2H}$	-0.73425
$E_{ads}(H_2O_2)_{Ag\text{-}2H}$	-0.71338
$E_{ads}(C_2H_5OH)_{Ag\text{-}2H}$	-0.77370

While 2D Au-2H nanosheet adsorbs the most hydrogen peroxide while 2D Ag-2H adsorbs it the least. The same goes for ethanol molecule that the ethanol molecule is adsorbed the most by the 2D Ag-2H but the least by 2D Au-2H.

To calculate the electronic properties and physicochemical properties, the energy values of HOMO and LUMO are measured, presented in Tab. 2. The sign of ΔEg shows the increase or decrease in Eg, so we employed the |ΔEg| absolute value.

Table 2 HOMO (E_H), LUMO (E_L) and Fermi (E_F) energy values (in eV) of H_2O, H_2O_2, and C_2H_5OH on 2D Au_{2H} and 2D Ag_{2H} nanosheets.

Structure	Au_{2H}	Ag_{2H}	Au_{2H}-H_2O	Ag_{2H}-H_2O	Au_{2H}-H_2O_2	Ag_{2H}-H_2O_2	Au_{2H}-C_2H_5OH	Ag_{2H}-C_2H_5OH
E_H	-3.7717	-3.2647	-3.5807	-3.4959	-4.1865	-3.1030	-3.2254	-2.9421
E_L	-3.1882	-2.4462	-2.3489	-2.3883	-3.1613	-3.0004	-1.9761	-1.9842
E_g	0.5835	0.8185	1.2318	1.1076	1.0252	0.1026	1.2493	1.5293
$\%\Delta E_g$	-	-	111.1054	35.3207	75.6984	**-87.4649**	114.4045	86.8418
E_F	-3.4799	-2.8554	-2.9648	-2.9421	-3.6739	-3.0517	-2.6007	-2.7488

Table 3 Physicochemical properties of H_2O, H_2O_2, and C_2H_5OH on 2D Au_{2H} and 2D Ag_{2H} nanosheets.

Structure	Au_{2H}	Ag_{2H}	Au_{2H}-H_2O	Ag_{2H}-H_2O	Au_{2H}-H_2O_2	Ag_{2H}-H_2O_2	Au_{2H}-C_2H_5OH	Ag_{2H}-C_2H_5OH
μ	-3.47995	-2.8554	-2.96480	-2.9421	-3.67390	-3.0517	-2.60075	-2.7488
S	1.71380	1.22175	0.81182	0.9028	0.97542	9.7466	0.80045	0.6539
η	0.29175	0.40925	0.61590	0.5538	0.51260	**0.0513**	0.62465	0.7446
ω	20.75416	9.96163	7.13593	7.8150	13.16576	90.7687	5.41415	4.9409
ΔE_e	24.37998	13.02170	10.40868	11.0340	17.09596	93.8470	8.32722	8.07211
ΔE_n	17.42008	7.3108	4.47908	5.1498	9.74816	87.7427	3.12573	2.5744
ΔN_{max}	11.92785	6.9773	4.81377	5.3126	7.16719	59.4873	4.16353	3.5949

Although the minimal adsorption of ethanol by 2D Au-2H nanosheet, it caused the largest enhancer band gap to change on Au_{2H}-C_2H_5OH. The band gap of complex Ag_{2H}-H_2O_2 decreased after the adsorption changed the band gap in a negative way. The band gap of Ag_{2H}-C_2H_5OH is less than the isolated 2H Ag nanosheet's.

The conductivity shown in Tab. 4 decreased significantly for both of nanosheets after the adsorption. The sensitivity depends on the electrical conductivity of the material.

Table 4 The electrical conductivity (σ) of H_2O, H_2O_2, C_2H_5OH on 2D Au_{2H} and Ag_{2H} nanosheets

Structure	σ	Sensitivity
Au-2H	1.1641×10^{-5}	-
Ag-2H	1.1991×10^{-7}	-
Au-2H-H_2O	3.8374×10^{-11}	99.999
Ag-2H-H_2O	4.3078×10^{-10}	99.641
Au-2H-H_2O_2	2.1430×10^{-9}	99.982
Ag-2H-H_2O_2	4.3078×10^{-10}	99.641
Au-2H-C_2H_5OH	2.7293×10^{-11}	99.999
Ag-2H-C_2H_5OH	1.1706×10^{-13}	99.999

We can express the deformation or change rate of complex structure with chemical hardness (η). The value chemical hardness of Ag-2H-C_2H_5OH is reduced despite the fact the values of other complexes are increased.

In order to use the 2D nanosheets as a means in biomedical applications, it is crucial to ensure that the molecule can be captured and retained by the surface for a very long time.

Table 5 Recovery time (τ) of H_2O, H_2O_2, C_2H_5OH on 2D Au_{2H} and Ag_{2H} nanosheets

Structure	$\tau^{UV\ vacuum\ light}$	$\tau^{UV\ radiation}$
Au-2H-H_2O	7.8539×10^{28}	2.1942×10^{25}
Ag-2H-H_2O	0.8716	0.0002
Au-2H-H_2O_2	1.3735×10^{29}	2.9608×10^{24}
Ag-2H-H_2O_2	0.3867	9.6642×10^{-5}
Au-2H-C_2H_5OH	1.8290×10^{28}	1.2593×10^{25}
Ag-2H-C_2H_5OH	4.0505	0.0010

The calculated recovery time are too small to desorb H_2O, H_2O_2, C_2H_5OH from Ag-2H nanosheet. Nonetheless, the recovery times for Au-2H nanosheet is too long (~10^{21} years).

IV. CONCLUSION

In this study, DFT calculations are performed to determine the electronic structure properties and the dynamics of H_2O, H_2O_2 and C_2H_5OH placed on the top of the 2D Au-2H and 2D Ag-2H nanosheets. The nanosheets with molecules after the optimization show us that the structure of 2H nanosheets turn into the more stable face-centered cubic structure. 2D Au-

2H hcp structure doesn't seem to change because the $(111)_{fcc}$ correspond with $(0001)_{hcp}$ surface. Negative adsorption energy suggests that the adsorption process of small molecules is exothermic. Also, the band gap of all complexes except Ag_{2H}-H_2O_2 increased. Au-2H nanosheet can be a candidate for possible applications for viral capture.

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