

CO Gas Adsorption on SnO₂ Surfaces: Density Functional Theory Study

* **Hayk Zakaryan and Vladimir Aroutiounian**

Yerevan State University, 1 Alex Manoogian, 0025, Armenia

Tel.: +37410555240, fax: +37410554641

*E-mail: zhayk91@gmail.com

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Abstract: This research is devoted to the investigation of the toxic carbon monoxide gas adsorption mechanisms on the tin dioxide semiconductor. We used density functional theory to describe adsorption processes and found out that the Mars-van Krevelen adsorption mechanism is not responsible for adsorption on (101) and (001) surface orientations of tin dioxide. For (110) and (100) surfaces, after adsorption carbon dioxide molecule forms and desorbs from the surfaces. For (101) surface orientation, carbon monoxide adsorb to the surface's oxide by carbon atom and stay bonded to it. Charge transfer from the molecule to the surface, which equal to 1.9e calculated by Bader charge analysis. In the case of (001) surface orientation, carbon monoxide adsorb to surface's oxygen and stay bonded too. Here, we consider half and full surface coverages. It was shown, that during full surface coverage, only one molecule can adsorb and transfer 2e charge. Electronic density of states calculation was done to explain the increase of surface conductance.

Keywords: Gas, Sensor, DFT, CO, Adsorption, Mars-van Krevelen.

1. Introduction

Carbon monoxide (CO) is tasteless and transparent substance, which is known as an "invisible killer" due to the high level of toxicity. It is extremely poisonous and can even cause health effects up to a certain extent – 9 ppm, therefore, the detection of CO in the environment is quite vital. For the above reasons, chemical sensors are used to precisely monitor the concentration of the target compounds in the air. The main part of chemical sensors is sensing or active material, which interact with gas molecules. Usually, for sensing materials are used metal oxides like TiO₂, SnO₂, ZnO, In₂O₃.

In modern world, where technology is developing every day, it is crucial to have precise and functional devices. For sensor technology, it means to have a sensing material, which has different high parameters

like high conductance, wide band gap, and high charge carriers' mobility. All these parameters have the SnO₂ semiconductor. The gas sensors made by it, has high sensitivity and low response time for wide variety of molecules, such as CO, H₂, CH₃OH, NO_x, etc. [1-8]. Sensitivity is the one of the most important parameters of sensors; it defines how much signal change when sensors detect the target gas. In order to increase the sensitivity of the detectors, it is crucial to deeply investigate adsorption mechanism of CO compounds.

Several models, which are named as Langmuir Hishelwood [10-11], Eley Redial [12] and the MvK [13], have been developed to describe the mechanisms that are responsible for CO adsorption on oxide semiconductors. The MvK adsorption mechanism is in a good agreement with the experiments on SnO₂–(110) surface [14-15]. Apparently, the MvK mechanism consists of following steps:

1) Adsorbed CO molecule is reacting with tin dioxide oxygen and forms CO₂ compound, leaving oxygen vacancy in the material;

2) Then remained vacancy fills by adsorbed O₂ molecule from the environment;

3) And finally, another CO molecule is reacting with already bonded O₂ and forms CO₂, leaving material in its initial undisturbed state.

We would like to stress that there are plenty of manuscripts dedicated to the investigation of CO absorption on SnO₂ surfaces, using first principle and ab initio DFT calculations [16-17]. Some calculations were done particularly for pristine (110) surface orientation [18], oxygen rich [15] and tainted surfaces [16]. However, very few are devoted to other surface orientations (100), (101) [5] and (001) [6]. In practice, surface orientation immensely influences sensor parameters such as sensitivity, time response etc.

In this manuscript, we explore CO adsorption on various surface orientations (110), (100), (101), (001) (Fig. 1) of SnO₂ and determine the most optimal configurations for adsorption, using ab initio DFT calculations in order to shed the light on the atomic scale processes that still remains elusive and unclear.

Here, we will rise following issues:

1) Does MvK mechanism similarly describing adsorption processes for all surface orientations of SnO₂?

2) What is the exact amount of charge transferred between the surface and adsorbed CO molecule?

3) Which surface orientation is more prominent to the interaction with the CO molecule?

The paper is organized in the following way: Section 2 is devoted to computational methods and surface/bulk structure of SnO₂ sensing material. Section 3 is devoted to the adsorption processes, eDoS calculations for the various surface orientations of SnO₂. Section 4 is dedicated to discussions and conclusions.

2. Models and Computational Methods

Calculations were done using conventional ab initio DFT [21-22] method implemented in Vienna ab initio Simulation Package (VASP) [23-25]. DFT relaxations were done within Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) [26]. The 500 eV cutoff energy was chosen due to the total energy convergence from that value. Surface structures were relaxed until the threshold net force on atoms become less than 0.01 eV/Å. The Monkhorst-Pack scheme [27] was used to sample the Brillouin zone, using 6×6×1 k-points mesh. During the calculations of adsorbed molecules on surface, each configuration was relax to find the most relaxed structure or the structure were the forces on atoms are the minimum. The ionic relaxation was done using conjugate gradient method implemented in VASP. After that relaxation, the Bader charge analysis was done to find out charge distribution around each atom

[28-30]. The difference, between charge distribution of the relaxed surface and pure surface, will be the amount of charge transferred to the surface after adsorption finished.

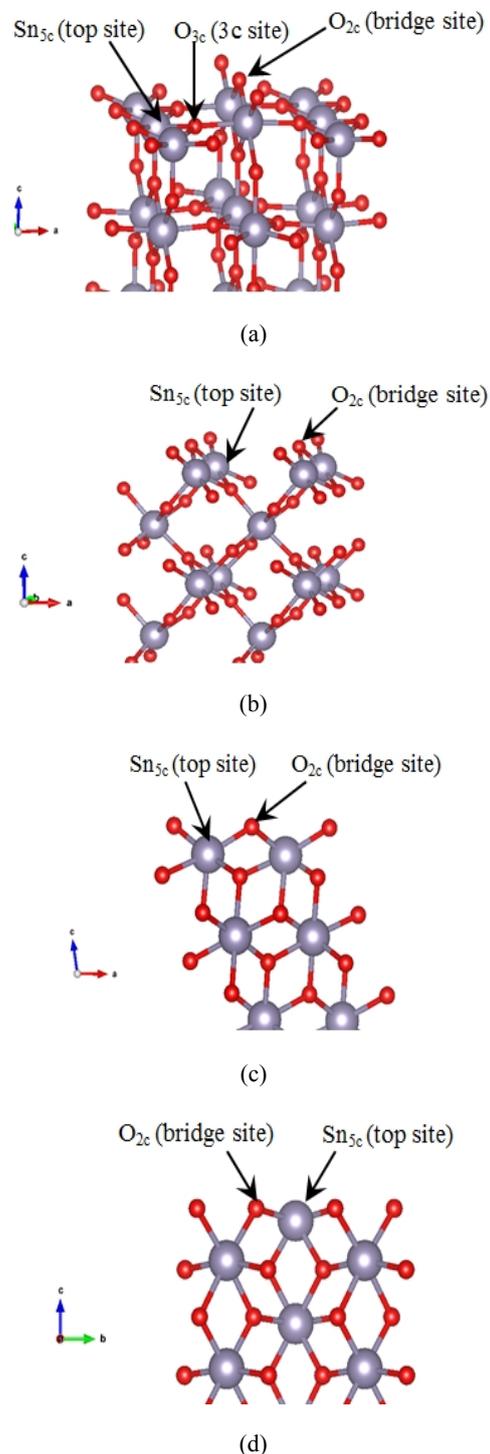


Fig. 1. Different surfaces of SnO₂ and possible adsorption sites on it. (a) - (110), (b) - (100), (c) - (101), (d) - (001).

Stable adsorbed configurations were found using the following equation:

$$E_{surf} = E_{ads} - E_{clean} - E_{CO}, \quad (1)$$

where E_{ads} is the adsorption energy, E_{surf} is the total energy of SnO₂ surface and adsorbed CO, E_{clean} is the total energy of pure surface without CO and E_{CO} is the total energy of the gas molecule. It is clear from this equation that if E_{ads} is negative the configuration of adsorbed site is stable, in other words, the process is exothermic. In the case of positive E_{ads} , the process is endothermic: the molecule will not adsorb to the surface and will remain in the non-interacting state.

Bulk SnO₂ has rutile, tetragonal structure, corresponding to the P42/mnm space group. The lattice parameters of SnO₂ from [31-32] are $a = 4.82 \text{ \AA}$, $c = 3.23 \text{ \AA}$ and $u = 0.307$. In our calculations (see Fig. 1) each (110), (100), (101), (001) surface consists of 4 layers and relative stability has the following sequence (110), (100), (101), (001) [6]. Here, number of layers was tuned to check the convergence of surface energy. However, after getting all results, we double checked the obtained data by recalculating stable structures with a big substrate of 12 atoms of tin and 24 atoms of oxygen and make sure that the results are reliable. For all calculations, we choose vacuum thickness of 15 \AA , which is greater than substrate thickness. For each surface, the electronic density of states (eDoS) was calculated and established that the gap between valence and conduction bands was underestimated, because of the self-consistency of DFT calculations. From experiments, it is known that value of band gap is 3.6 eV, but we get 1 eV, which is in accordance with previous DFT calculations [31]. Despite of the gap underestimation, it doesn't influence on the value of the adsorption energies and bond lengths of the calculation. Even the gap is narrower than in experiment; the trend of band gap change after gas molecule adsorption would be the same, because charge transfer or charge density calculated by PBE has minimal error between known functionals.

3. Results and Discussion

In this section, each surface orientation would be described separately. Bader charge analysis, eDoS and stable structures would be presented as well.

3.1. SnO₂ (110) Surface

This surface orientation consists of 4 layers, each comprising of 3 subsequent layers. Top sublayer represents oxygen, where each atom (O_{2c}) is connected by the covalent bond to 2 Sn atoms. The Second sublayer has 2 Sn atoms and 2 oxygen atoms. Third one is similar to the first layer. So, together these 3 sublayers can be treated as one layer which is continually repeated 4 times (see Fig. 1(a)). There are 3 possible adsorption sites: top site (t), bridge site (br), three-coordinated oxygen site (3c). For each site, we consider 2 configurations:

1) C atom in a CO molecule is closer to the surface (C down configuration),

2) The vice versa configuration (O down configuration). Thus, we end up with 6 possible configurations for (110) surface.

For each separate configuration calculated adsorption energies are given in Table 1. As we can clearly see the O down configuration for all surfaces is completely unstable.

In the case of C down configuration, (110) surface has 3 stable sites. However, there is only one that is transferring the charge from molecule to the surface, and it is corresponding to the event when molecule approaches to br-site, reacts with oxygen and takes it away forming a CO₂ molecule and leaves oxygen vacancy on the surface. Here, vacancy could be an adsorption site for O₂ or CO molecule, as described in [16, 19]. The eDoS of surface reveals that conductivity of the surface increases by decreasing of the band gap. The Bader charge analysis shows charge transport to the surface of 1.7e. Such processes examined in a number of experimental and theoretical studies [14], [16, 19]. It is important to note, that for this particular configuration and surface orientation the first step of MvK mechanism is preserved, and the distance between CO₂ molecule and surface is 3.1 \AA .

Table 1. Adsorption energies, distances and bader charge analysis of (110) surface.

Ads. sites	E_{ads} (eV)	CO distance d surface (\AA)	Charge transferred to surface (e)
br	-0.48	3.1	1.7
br (O down)	0.31	2.58	0.0
top	-0.16	2.465	0.0
top (O down)	0.01	2.69	0.0
3c	-0.03	2.61	0.0
3c (O down)	0.5	2.62	0.0

For other sites (top and 3c) the CO molecule does not exchange electrons, which serves as a confirmation that we have physisorption process onto the surface. In these cases, band gap does not change and distances from carbon to tin atoms are 2.46, 2.61 \AA , respectively

3.2. SnO₂ (100) Surface

For (100) surface orientation the unit cell also consists of same 4 layers, where each layer can be divided into 3 sublayers: O-Sn-O layers. The top atom of the surface is 2 coordinated oxygen as in (110) surface, see Fig. 1(b). Adsorption energies for possible two site configurations are given in Table 2. According to the calculations interaction of CO with the O_{2c} site of oxygen leads to the formation of CO₂

molecule and also leaves a vacancy on the surface (Fig. 2(a)).

Table 2. Adsorption energies, distances and bader charge analysis of (100) surface.

Ads. sites	E_{ads} (eV)	CO distanced surface (Å)	Charge transferred to surface (e)
br	-0.48	3.25	1.6
top	-0.16	2.65	0.0

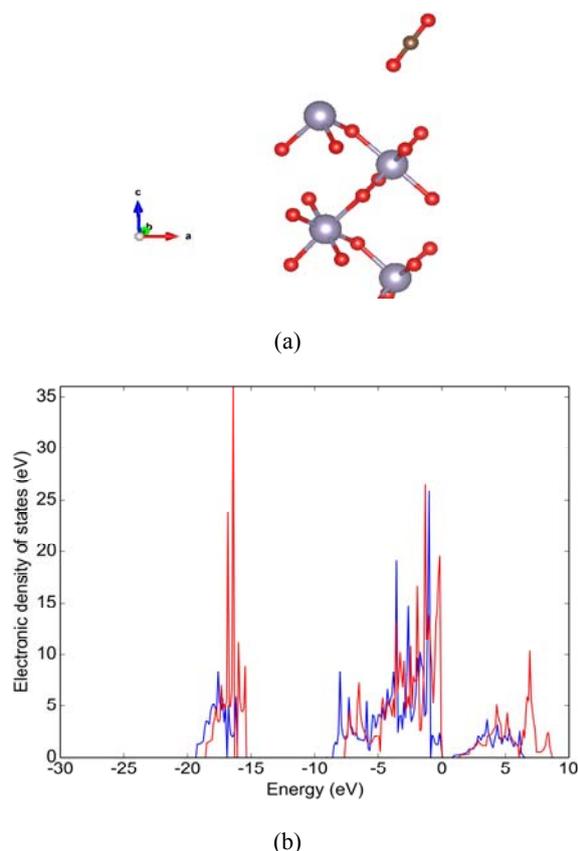


Fig. 2. Structure and eDoS of (100) surface. (a) Desorbed CO_2 molecule from (100) surface (red circle is oxygen, grey is Sn, brown is C atoms). (b) eDoS of substrate before adsorption (red line), and after CO_2 desorption (blue line).

In this case, the first step of MvK mechanism is also conserved and it leads to charge transfer of 1.6e to the surface. Therefore, the surface conductance is increasing, because of transferred charge leading to the band gap reduction by 0.1 eV in comparison to the uncharged surface, see Fig. 2(b). Here, the blue lines represent the eDoS after desorption that correspond to 3.25 Å distance from CO molecule to surface.

The Second possible adsorption site is top (t) on Sn atom. Here as for (110), the CO is physisorbed and no charge transfer has been observed. Due to physisorption, the distance from a carbon atom of CO molecule to the oxygen atom of the surface is 2.65 Å.

3.3. SnO_2 (101) Surface

In the case of (101) surface orientation, each of 4 layers consists of 3 subsequent layers of 2O, 2Sn, 2O. There are 3 possible sites of adsorptions O_{2c} , Sn atom and 3 coordinated O, which is located in third sub layer as it is shown in Fig. 1(c). Here, only one configuration has negative adsorption energy and it is O_{2c} (Table 3).

Table 3. Adsorption energies, distances and bader charge analysis of (101) surface.

Ads. sites	E_{ads} (eV)	CO distanced surface (Å)	Charge transferred to surface (e)
O_{2c}	-0.47	1.18	1.9

The CO molecule adsorbs by the surface and remains connected to it in O_{2c} site with a distance of 1.18 Å (see Fig. 3). During that process, the distance between O_{2c} oxygen and Sn atom increases up to 2.5 Å. The Bader charge analysis shows that there is 1.9e charge transferred to the surface, which make it more conductive. Thus, adsorption mechanism on (101) surface differs from MvK, because no CO_2 desorption observed. Conductivity increases and band gap decreases due to transferred charge, see Fig. 3(b), where blue lines represent eDoS after adsorption and have more states around Fermi level (Energy = 0 in the horizontal axis). To find the whole adsorption mechanism it is necessary to do molecular dynamics simulation (MD), which should be done in future researches.

3.4. SnO_2 (001) Surface

In this case, we have a completely different situation. Instead of 4 layers that consist of 3 subsequent layers, we have got only one that consists of one atom of Sn and 2 oxygen atoms. There are two possible sites: on top of Sn (t) site and O_{2c} (2c) site Fig. 1 (d). Moreover, there are 2 equivalent 2c sites in one unit cell, thus we should take into account two possible coverages. The first is when both sites are occupied by the CO molecule and form one mono layer (ML=1). The second possible coverage is when only one site is occupied and half mono layer of CO forms (ML = 0.5).

For ML = 0.5, carbon monoxide adsorbed and stay bounded with a distance of 1.16 Å (Fig. 4) to the O_{2c} atom, transferring 2e charge to the substrate (see Table 4).

For ML = 1, one CO molecule adsorbed and one physisorbed in a 1.255 and 2.6 Å distances, respectively. In fact, for (001) only ML=0.5 coverage can happen, because when one CO adsorbed, the

second one will be physisorbed. Bader charge analysis shows that 1.83e was transferred to the surface.

In both cases, surface conduction increases and band gap decreases due to charge transfer from the CO molecule.

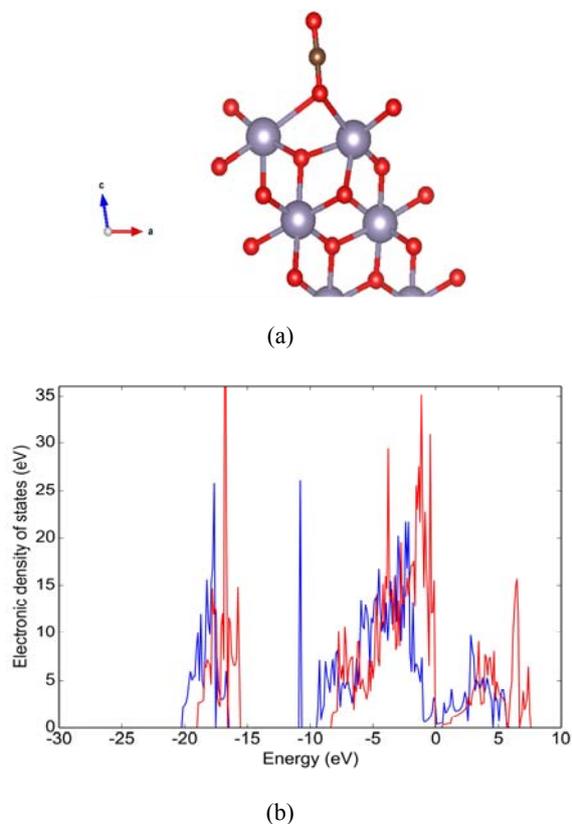


Fig. 3. (a) Adsorbed CO on (101) surface (red circle is oxygen, grey is Sn, brown is C atoms). (b) eDoS of clean (101) surface (red line) and CO adsorbed on it (blue line).

Table 4. Adsorption energies, distances and bader charge analysis of (001) surface.

Ads. sites	E_{ads} (eV)	CO distance d surface (Å)	Charge transferred to surface (e)
2c, ML = 0.5	-1.19	1.16	2.0
2c, ML = 1	-1.24	1.25	1.8
top	-0.19	2.41	0.0

On Fig. 4 (b)(d), the red lines represent eDoS of undistorted surface and blue correspond to the eDoS after adsorption, which has more states around Fermi level compared to undistorted surfaces eDoS. Physisorption occurs for the top site with distance 2.41 Å.

So, MvK mechanism is not responsible for adsorption on (001) surface orientation, because CO molecule adsorb and remain bonded to the surface instead of forming CO₂.

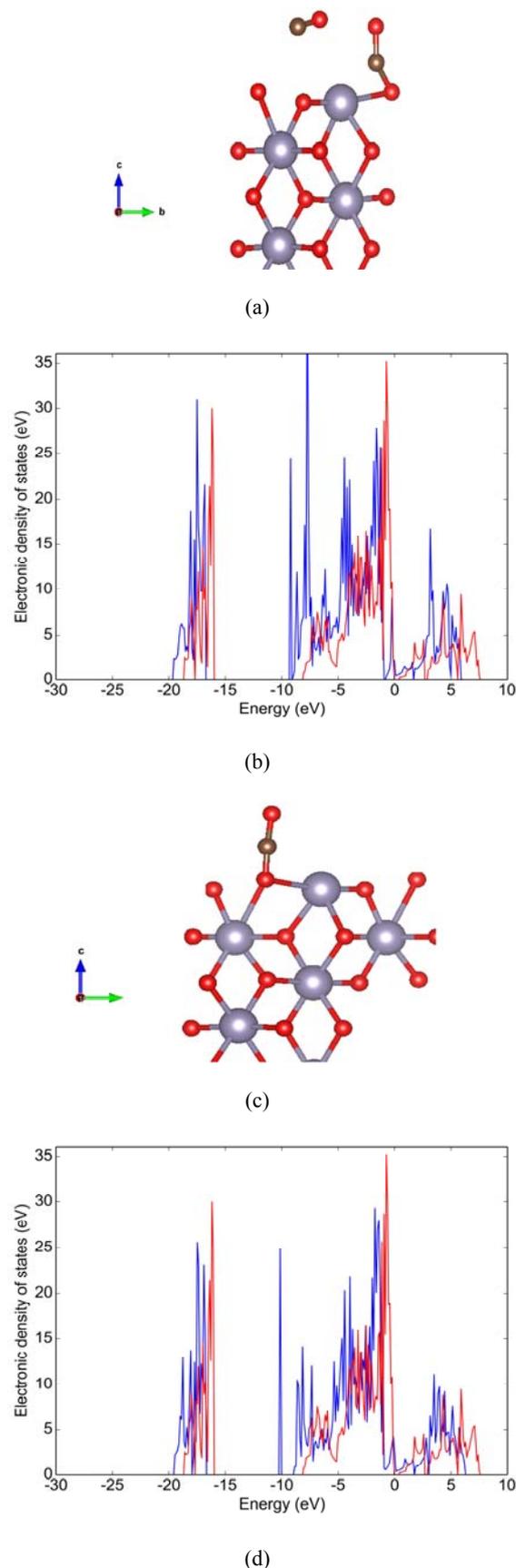


Fig. 4. Configurations and eDoS of (001) surface (red circle is oxygen, grey is Sn, brown is C atoms). (a) Adsorption when ML=1. (b) eDoS before and after adsorption, when ML = 1. (c) Adsorption when ML= 0.5. (d) eDoS before and after adsorption, when ML = 1.

4. Conclusions

Previously, it was considered that adsorption on SnO₂ surfaces follows MvK mechanism; however, here we have proven that CO adsorption on SnO₂ (101), (001) is different. Here, the C atom of the CO molecule remains bonded to surface's O atom. Also, we find out that in the case of (001) surface orientation CO coverage can be only half mono layer (ML = 0.5). For (110) and (100) surfaces, we establish that adsorption obeys MvK mechanism, where its first stage CO₂ molecules are forming during CO-surface interaction. For all adsorption cases, O down configuration was not stable, due to positive adsorption energy. To find whole adsorption mechanism for (101) and (001) surface orientations, it is necessary to do MD calculation and find next steps of reaction between CO and surface. This will be done in the future research.

The Bader charge analysis reveals that charge transfer to (101), (001) surfaces are 1.9e, 2e respectively and 1.7e, 1.6e for (110), (100) surfaces. The eDoS combined with Bader analysis shows that (101), (001) surface orientations gather more electrons than the rest orientations, thus, those should be considered as a better platform for the interaction of the CO molecules with SnO₂ surfaces. We believe that our findings will pave the way for the fabrication of SnO₂ based CO sensors with higher sensitivity and lower response time.

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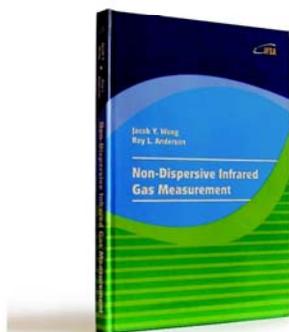
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