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Title: Observation of the electronic states from Au/TiO₂ (320) interface as a function of the azimuthal angle and polarization by optical second harmonic generation

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Abstract:

The Au/TiO₂ interface shows remarkable catalytic activity for many chemical reactions especially for the oxidation of CO and cracking of H₂O. Study of the electronic state of the Au/TiO₂ (320) interface is vital to explore the mechanism of the catalytic behaviour. In order to contribute to the sector of catalyst chemistry, we observed the electronic states of Au/TiO₂ (320) interface using second harmonic generation (SHG) method. SHG is a well-established surface-specific probe of centrosymmetric media. In the dipole approximation, SHG is forbidden in the bulk of a medium having inversion symmetry, while at the surface inversion symmetry is broken and SHG is allowed. Au/TiO₂ steps should generate a SHG signal due to broken symmetry at the interface. The main aim in this study is to detect this signal. We fabricated an Au thin film with the thickness of 2 nm on a stepped TiO₂ (320) substrate in a UHV chamber at a pressure of 2×10^{-7} Torr. We observed the SHG intensity as a functions of the azimuthal angle and polarization from Au/TiO₂ (320) interface and bare TiO₂ (320) using both 1064 nm and 532 nm wavelength of pulsed Nd²⁺:YAG laser light. When using 1064 nm, we found isotropic responses from both samples. We found anisotropic response from both Au /TiO₂ (320) and bare TiO₂ (320) by using 532 nm of laser light as the incident light. For Au deposited TiO₂ (320) sample, the Pin-Pout SHG pattern showed an anisotropy to the $[2\bar{3}0]$ direction. We decomposed theoretically the optical nonlinear susceptibility elements $\chi_{ijk}(2)$ into odd and even order group. Here, i, j and k denote the axis direction. The odd order group contain the odd number of 2 in the $\chi(2)$ elements of this group. Here, 2 is the direction of the step of the sample. In the similar way, we define even order group which contain odd number of 3 in the $\chi(2)$ elements of this group. Here, 3 is the terrace direction of the sample. This anisotropic response of Pin-Pout for Au deposited TiO₂ (320) sample contains the contribution of Pin-Pout configuration of odd order and even order group $\chi_{ijk}(2)$ element (shown in the below diagram). We only observed the anisotropic response when using 532 nm as incident probe from the stepped Au/TiO₂ (320) interface, so the electronic resonance of the Au covered step is detected in the ultraviolet region particularly at 266 nm of SHG signal. This electronic resonance may be responsible for many catalytic reactions.