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Title: The Au/TiO₂ (320) interface study by using optical second harmonic generation technique

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Published Conference Name: Japan-India Symposium on Materials Science 2017

Type of Publication: International Conference

Volume: _____ **Issue** _____

Publisher: _____

Publication Date: March 2017

ISSN: _____

DOI: _____

URL: _____

Other Related Info.: _____



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Abstract:

The Au/TiO₂ interface is very fascinating to study in catalyst point of view. Studying the electronic states of the Au/TiO₂ interface is vital to explore the catalytic mechanism. Many researchers already studied the Au/TiO₂ (110) interface by several optical microscopic technique such as scanning tunnelling microscope (STM), transmission electron microscope (TEM), UV-Vis spectroscopy, and so on, but the number of studies of Au/TiO₂ (110) interface by second harmonic generation (SHG) is very limited. So we intended to study the Au/TiO₂ (320) interface by SHG technique for the first time. For developing and enriching the catalyst field, we observed the Au/TiO₂ (320) interface to explore their electronic states by using SHG technique. 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 origin of the second order nonlinearity is the breaking of symmetry in the direction normal to the surface and also direction parallel to the surface and this may occur due to the discontinuity, reconstruction or relaxation on the surface. The Au film was deposited in a UHV chamber at a pressure of 2×10^{-7} Torr. The approximate thickness of Au film on a stepped TiO₂ (320) substrate surface was 2 nm. We observed the SHG response from the Au/TiO₂ (320) interface and bare TiO₂ (320) using both 1064 nm and 532 nm wavelength of pulsed Nd²⁺:YAG laser light. The isotropic response was found from both samples in case of using 1064 nm wavelength of laser light. In contrast, we observed the anisotropic response from both Au /TiO₂ (320) and bare TiO₂ (320) when we used 532 nm of laser light as the incident probe. From Au/TiO₂ (320) sample, an anisotropic structure was observed to the $[2\bar{3}0]$ direction for Pin/Pout polarization combination. We theoretically decomposed the nonlinear susceptibility elements ($\chi_{ijk}(2)$) and divided them into two groups such as step contribution and terrace contribution groups. Here, i, j and k denote the axis direction of the sample coordinate. The anisotropic response was only observed for wavelength 532 nm from the stepped Au/TiO₂ (320) interface. In conclusion, we infer that a resonance between the electronic states of the Au covered step with the energy of the 4.66 eV near UV second harmonic photon may be responsible for the signal, and that these electronic states may be responsible for the catalytic activity of Au/TiO₂.