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Title: Investigation of the Au/TiO₂ (320) Interface by Optical Second Harmonic Generation Technique

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Abstract:

For many catalytic reactions, the Au/TiO₂ interface acts as an active site. It exhibits an extraordinary high activity for low-temperature catalytic combustion, partial oxidation of hydrocarbons, hydrogenation of unsaturated hydrocarbons, and reduction of nitrogen oxides and so on. In order to contribute to the understanding of the catalyst, we investigate the electronic state of the Au/TiO₂ (320) interface by second harmonic generation (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. As the surface steps show noncentrosymmetric behavior, Au/TiO₂ steps should generate a SHG signal. The target of this study is to detect this signal. We fabricated an Au thin film on a stepped TiO₂ (320) substrate in a UHV chamber at a pressure of 2×10^{-7} Torr. The thickness of the film was 2 nm. The azimuthal angle and polarization dependent SHG intensity from Au/TiO₂ (320) interface and bare TiO₂ (320) were observed by using both 1064 nm and 532 nm wavelength pulsed laser light. When using 1064 nm, we found isotropic response from both the Au/TiO₂ (320) interface and bare TiO₂ (320). This behavior is dominated by the $\chi_{311}^{(2)}$ and $\chi_{322}^{(2)}$ nonlinear susceptibility elements. Here, 1, 2 and 3 represent $[2\bar{3}0]$, $[001]$ and $[320]$ directions respectively. When using 532 nm, we found anisotropic behavior from both Au/TiO₂ (320) and bare TiO₂ (320). For Au deposited TiO₂ (320) sample, the P_{in} - P_{out} SHG pattern showed clear anisotropy to the $[2\bar{3}0]$ direction. The bare TiO₂ (320) sample also revealed anisotropy. This anisotropic behavior is dominated by $\chi_{113}^{(2)}$ nonlinear susceptibility element. The anisotropic behavior was only found using wavelength 532 nm as incident light. The SHG intensity patterns from Au/TiO₂ (320) interface and bare TiO₂ (320) surface had different anisotropic behavior and their dominating nonlinear susceptibility element $\chi^{(2)}$ were also different. It is thought that a resonance between the electronic states of the Au covered step with the energy of the 266 nm near UV second harmonic photon may be responsible for our observations, and that these electronic states may be responsible for the catalytic activity of Au/TiO₂.