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

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Abstract:

The Au/TiO₂ interface acts as an active site for many catalytic reactions such as reduction of nitrogen oxides, partial oxidation of hydrocarbons, hydrogenation of unsaturated hydrocarbons, oxidation of carbon monoxides and so on. In order to contribute to catalyst field, we investigate the electronic state of Au/TiO₂ (320) interface by second harmonic generation (SHG) technique. SHG is a surface-active well-established phenomenon especially for noncentrosymmetric 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 shown on centrosymmetric behavior, Au/TiO₂ steps should generate a SHG signal. We fabricated a Au thin film on the stepped TiO₂ (320) substrate in a UHV chamber at 2×10^{-7} Torr with the film thickness of 2 nm. We observed the azimuthal angle and polarization dependent SHG intensity from the Au/TiO₂ (320) interface and bare TiO₂ (320) using both 1064 nm and 532 nm wavelength of pulsed laser light as fundamental source. In case of wavelength 1064 nm, we found isotropic responses from both samples. This behavior is dominated by the $\chi_{311}(2)$ and $\chi_{322}(2)$ nonlinear susceptibility elements. Here 1, 2 and 3 represent [230], [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 Pin-Pout SHG pattern showed clear anisotropy in the [230] 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 the Au/TiO₂(320) interface and bare TiO₂ (320) surface had different anisotropic behavior and their dominating nonlinear susceptibility element $\chi(2)$ were also different. As we observed the anisotropic response at 532 nm fundamental light from the stepped Au/TiO₂ (320) interface, the electronic resonance of the Au covered step is detected in the ultra-violet region particularly at around wavelength 266 nm as SHG signal. This electronic resonance may be responsible for many catalytic reactions.