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Title: Azimuthal angle and polarization dependent second harmonic generation investigation from Au/TiO₂ (320) interface

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Abstract:

Au/TiO₂ shows an extraordinary catalytic activity such as partial oxidation of hydrocarbons, oxidation of carbon monoxides, hydrogenation of unsaturated hydrocarbons, reduction of nitrogen oxides and so on. In order to understand the catalytic mechanism, the study of electronic states of the Au/TiO₂ interface is important. Thus, our intension was to study electronic states of the Au/TiO₂ (320) interface by second harmonic generation (SHG) technique. SHG is a well-established surface-specific probe of non-centrosymmetric media. In the dipole approximation, SHG is forbidden in the bulk of a medium having inversion symmetry but allowed at the surface and interface where inversion symmetry is broken. Au/TiO₂ sample has a stepped structure at the interface so that it should generate SHG signals due to broken symmetry. In this research, Au film was deposited on the stepped TiO₂ (320) substrate with the thickness of 2 nm in a UHV chamber at a pressure of 2x10⁻⁷ Torr. We investigated the SHG response from the Au/TiO₂ (320) interface and bare stepped TiO₂ (320) using pulsed Nd²⁺:YAG laser light at the fixed photon energies of 1.17 eV and 2.33 eV individually. At the photon energies of 1.17 eV, almost isotropic responses were found for all polarization combinations from both samples. However, anisotropic responses were observed when we measured the same polarization combinations at the photon energy of 2.33 eV. Especially, an intense anisotropic response with heart shape was observed in the [2̄30] direction for Pin/Pout polarization combination as shown in the left-hand side of a below figure. We theoretically decomposed the nonlinear susceptibility elements and divided them into two groups as step contribution and terrace contribution described in the following figure. The interesting point in this research is the separation of step contribution in the SHG signal of interface which could not be done by other methods. This result is promising to find the actual resonance band by scanning the excitation wavelengths, from which the origin of catalytic mechanism of the stepped Au/TiO₂ interface can be understood.