Scanning non-linear optical probe microscopy utilizing tip-enhanced near field optics

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Surfaces and interfaces among thin films of metals, metal oxides, water solutions, and organic materials are getting more and more important, recently, because of interesting scientific findings and important industrial applications. Investigating light-material interactions at a hetero interface is important also to understand physics of the materials for quantum devices. High spacial resolutions of scanning probe microscopy (SPM) enabled identifying properties of the materials interfaces that had not been otherwise accessible. In this work, we describe a novel SPM to observe non-linear optical properties of the interfaces at a 10 nm level spacial resolution.

We utilize sum-frequency generation (SFG), one of the second-order nonlinear optical processes which give signal intensity only where the spacial symmetry is broken. For example, we can investigate a 1.5 nm thick hydration structure of the water interface using SFG spectroscopy [1]. However, the spatial resolution of the traditional SFG microscopy (1 μm) is not enough for measuring nanometer-scale interface properties. Therefore, we combined SFG with SPM utilizing tip-enhanced near field optics [2].

We examined a gold thin film surface in air and measured the topography of the surface and SFG intensity of water molecules adsorbed on the surface. We observed local peaks of the SFG intensity where the topography showed local troughs. The results can be understood by a gold surface model with contaminations which give higher topographic height and lower SFG signal.

[1] Q. Du et al., Phys. Rev. Lett. 72, 238 (1994).
[2] N. Hayazawa, Y. Inouye, Z. Sekkat, and S. Kawata, Opt. Commun. 183, 333 (2000).