14-P7
SIMULTANEOUS MEASUREMENT OF AMPLITUDE AND PHASE IN SURFACE SHG

Jun Chen*, and Susumu Machida
Email: chen@photo.t-kougei.ac.jp
ERATO Yamamoto Quantum Fluctuation Project
NTT Musashino R&D Center, 3-9-11 Midoricho, Musashino, Tokyo 180, Japan

Recently, Optical second-harmonic generation (SHG) proved to be a sensitive and versatile probe of surfaces and interfaces [1]. It was demonstrated that one can use this technique to probe the structure and symmetry and electric properties of oxidized or reconstructed silicon surfaces. This technique was also used to determine the orientation of surface molecules and surface structure by measurement of the SHG phase. However, such applications have been restricted by the photon counting method used for SHG detection, because it is difficult to simultaneously measure the intensity and phase of SHG waves. In this study, we present an ac optical balanced homodyne detection (OBHD) system which enables us to simultaneously measure, with extremely high sensitivity and precision, both the intensity and phase of a surface SHG signal [2]. The OBHD system basically consists of a Mach-Zehnder interferometer. A mode-locked Kerr-lens Ti:sapphire laser (Coherent Mira 900) pumped by an Ar-ion laser is used as a light source that produces linearly polarized 150-fs pulses at a repetition of 76 MHz. The laser beam is split into the signal and local oscillator (LO) arms. In the signal arm, the laser beam is focused by a lens to an approximately 20 mm spot on the sample with an incident angle of 45° to generate the surface SHG signal. In the LO arm a 0.4-mm-thick BBO crystal is used to generate the LO wave. The signal and LO pulses are then temporally overlapped and yield two interference signals. The residual fundamentals in signal and LO arms interfere and yield interference signals too. Both the SHG and fundamental interference signals are then detected by photodiodes with a high quantum efficiency. The photodiode outputs are electrically subtracted and amplified using low-noise preamplifiers, respectively. The additive noise caused by the LO energy fluctuation can be reduced by balancing of the photocurrent from the two photodiodes. The optical path of the signal arm is modulated by a moving mirror. Such a modulation generates sinusoidal output signals at the amplifiers. The amplitude and phase of the SHG interference signal is then measured using a lock-in amplifier while the fundamental interference signal is used as a reference phase. The phases of the fundamental and SHG interference signals are strongly correlated, thus any phase shift caused by instability of the interferometer is automatically removed. By this method, the amplitude and phase in surface SHG can be simultaneously measured with extremely high sensitivity and precision. Several experiments were carried out using the SHG signal generated at the Si(111) surfaces. The sample used in our experiments was cut from a 0.35-mm-thick n-type Sb-doped silicon wafer with a resistivity of 0.1 W cm. The lowest SHG signal that can be detected using the developed system was estimated to be 3 aW corresponding to 6 photons/second. Simultaneous measurement of azimuthal rotation angle dependence of the intensity and phase in the p- and s-polarized SHG reflected from the Si(111) surface was demonstrated. This method makes the SHG technique more useful for in situ probing of surface dynamics and reactions.

[1] T. F. Heinz, M. M. T. Loy & W. A. Thompson, Phys. Rev. Lett. **54** 63 (1985).
[2] J. Chen, S. Machida &and Y. Yamamoto, Opt. Lett. **23** 676 (1998) .

*Present address: Faculty of Engineering, Tokyo Institute of Polytechnics, Atsugi, Kanagawa 243-0297, Japan.