

SECOND HARMONIC AND SUM-FREQUENCY GENERATION NONLINEAR OPTICAL ELLIPSOMETRY

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Second harmonic generation (SHG) and sum-frequency generation (SFG) have developed into remarkably powerful tools for characterizing surfaces, interfaces, and thin films because of their inherent surface specificity. The incorporation of full polarization analysis into these instruments will provide valuable insight into monolayer films of various chemical systems. The first-generation SHG instruments utilized physically rotating optical elements for polarization control and analysis. The techniques of nonlinear optical null ellipsometry (NONE) and rotating quarter-wave plate nonlinear optical ellipsometry (RQ-NOE) provide accurate and precise determination of the surface $\chi^{(2)}$ tensor elements. However, the long acquisition times required by these techniques limited their incorporation into an SFG spectrometer. The second generation SHG-NOE instrument utilized a discrete retardance (DR) configuration for the retarding optical elements. The DR-NOE system was specifically designed to perform NOE analysis while maximizing sample and application flexibility and minimizing data acquisition time. The DR-NOE technique for fast determination of the surface $\chi^{(2)}$ tensor elements was incorporated into a vibrational SFG spectrometer. The advantage of incorporating detailed ellipsometric measurements into a SFG spectrometer is the NOE measurements can assist in peak assignments and symmetry analysis in vibrational SFG spectroscopy measurements of surfaces and interfaces.