BROADENING THE HORIZONS OF NONLINEAR OPTICS BY NOSE

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Second harmonic generation (SHG) and has developed into powerful a tool for characterizing oriented thin films, surfaces, and interfaces. Furthermore, the nonlinear optical nature of the wave-mixing processes typically results in the generation of coherent exigent light with a well-defined polarization state. This coherence offers unique opportunities for extraction of detailed molecular and surface properties from polarization analysis. In previous studies, nonlinear optical ellipsometry (NOE) has been developed as a means to retain sign and phase information between the different nonzero $\chi^{(2)}$ tensor elements present in a given sample. However, those previous methods and related approaches for polarization analysis have all relied on the physical movement of optical elements in order to perform the analysis. The time required to physically move the appropriate optical elements ultimately dictates the fastest analysis time possible in a given technique. Such long acquisition times have limited NOE analysis to systems exhibiting excellent photostability. Development of Nonlinear Optical Stokes Ellipsometry (NOSE) has alleviated many of these problems. By increasing the repetition rate of the laser system and replacing previously slow rotating polarization optics with a rapid photoelastic modulator the acquisition time with full polarization analysis has been reduced from several hours to less than a second. This more than four order of magnitude reduction in acquisition time is accompanied by an order of magnitude more precision in $\chi^{(2)}$ tensors than previously achieved. These improvements have enabled imaging with full ellipsometric analysis at each pixel, allowing a unique contrast mechanism based on principle component analysis of the polarization dependent signal. Additionally insight into crystal quality and orientation of chiral crystals is available.