Differential ellipsometric surface plasmon resonance sensors with liquid crystal polarization modulators

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(Received 21 June 2004; accepted 17 August 2004)

Differential ellipsometric interrogation of surface plasmon (SP) resonances is a technique that gives ultrahigh sensitivity to refractive index changes, and it may provide the basis for chemical and biological sensors. In this study, a liquid crystal polarization modulator has been developed to provide such a differential technique. A refractive index sensitivity of 2×10^{-7} refractive index units is demonstrated, which is at least as sensitive as more established SP sensing techniques. The use of a liquid crystal modulator allows for low-voltage signal modulation and also feedback locking to zero. Possibly more important, it leads to pixelization for array sensing and for potential imaging. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1806273]

The use of optical chemical and biological sensors has seen much expansion in recent years, with a corresponding demand for improved transducers. One technique that could be utilized in ultrasensitive and robust sensors is that of differential surface plasmon ellipsometry.[1] However, the equipment generally used for this technique has so far not been low-voltage driven, and tends to be both bulky and expensive. In addition there is no provision in the technique for array sensing or multiplexing (allowing many sample areas to be monitored simultaneously or imaging). In this letter, a liquid crystal polarization modulator is developed that overcomes these difficulties, clearing the way for the technique to be developed further as a flexible refractive index (and hence chemical and biological) sensor.

It is well known that TM-polarized light may excite a surface plasmon resonance (SPR) at a metal/dielectric interface[2] in the Kretschmann configuration.[3] It has also been shown that there is a significant phase change of the reflected TM-polarized light as the SPR is traversed, whether by changing the incident angle, the wavelength of the light, or the refractive index of the bounding dielectric, and this has been utilized in sensors using heterodyne interferometry.[4,5]

If linearly polarized light consisting of both TM and TE polarizations is incident upon the SP system, then the TM-polarized component undergoes this phase change, whereas the TE-polarized component does not. The result of having two orthogonal components phase shifted with respect to each other is that the light reflected from the SP system becomes elliptically polarized. Because the phase changes rapidly as the SPR is traversed, the ellipticity and orientation of the polarization ellipse also changes rapidly. In this study, only the change in the azimuth of the polarization ellipse is considered since the variation in this is greater, as a function of the refractive index of the bounding dielectric medium, than is the ellipticity. Indeed, it can be shown through multilayer optics modeling[6] that the azimuth of the ellipse is rotated by approximately 1° for a refractive index change of only 5×10^{-5} refractive index units (RIU). Therefore, all that is needed to produce a sensitive refractive index sensor is a sensitive measure of the rotation of the polarization ellipse.

A polarizer placed in the path of an elliptically polarized beam, and rotated through an angle ϕ (relative to the major axis of the ellipse, produces a signal with a $\cos^2\phi$ dependence, with some amplitude reduction and constant offset due to the ellipticity. The angle at which the maximum in this dependence occurs corresponds to the azimuth of the polarization ellipse. If the plane of polarization of incident light upon a SP system is dithered sinusoidally, and the reflected signal monitored using a phase-sensitive detector with the reference set at the dither frequency, then the differential of the $\cos^2\phi$ curve is obtained as a function of the angle of the polarizer set in front of the detector. The zeros of this differential signal correspond to the azimuth or the azimuth $\pm\pi/2$ rad of the polarization ellipse. If the refractive index of the bounding dielectric medium is altered, the angular position of the zero in the differential signal also changes. For a more complete description of the method, see Ref. 1.

In the previously published paper,[1] a Faraday rotator produced dither of the polarization. From an applications viewpoint this has drawbacks. First, the Faraday rotator is relatively bulky and requires substantial power (in the magnetic coil) to produce the polarization dither. Second, the glass in the Faraday rotator is rather expensive, and third, the system cannot be easily pixelated to facilitate multiplexing.

Another way of producing the same effect is to use a liquid crystal (LC) cell, which is low-voltage driven, has low power consumption, and is cheap, small, and lightweight. Further, it also allows simple pixelization, for imaging or sampling many areas simultaneously. Here, a chiral hybrid aligned nematic LC cell is used.

A hybrid aligned nematic (HAN) LC cell has a director (the director describes the average orientation of the long molecular axis of the nematic liquid crystal) tilt, which changes almost linearly through the cell from homogeneous alignment (parallel to the cell substrate) on one surface to homeotropic alignment (perpendicular to the substrate) on the opposite surface. By adding a chiral dopant to the LC a twist of the director through the cell is produced, with the amount of twist being determined by the concentration of the dopant.

This twist produces a rotation in the plane of polarization of transmitted light through the cell. If a voltage is applied across the cell the LC director reorients and untwists to an extent dictated by the voltage (without a threshold as a HAN cell). Therefore, the amount of polarization rotation is controlled by the applied voltage.

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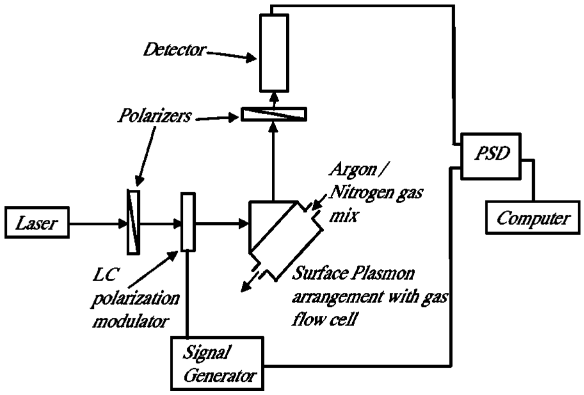

FIG. 1. A schematic of the experimental arrangement used.

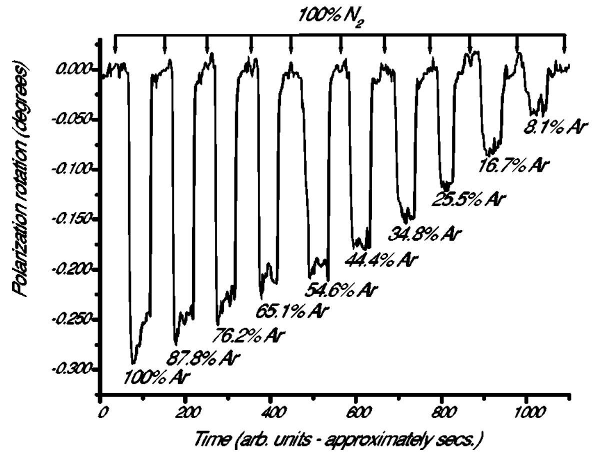

FIG. 2. The measured feedback signal (converted to a polarization rotation) obtained as the proportion of argon in the gas mix was changed.

To change the LC director distribution, a high-frequency (>10 kHz) sine wave is applied to the cell rather than a dc voltage to prevent electrostatic degradation of the LC (10 kHz is sufficiently high that the LC cannot respond to the fast oscillation, and it responds to the rms value of the voltage). In order to produce the desired polarization dither, amplitude modulation (500 Hz and 300 mVpp) of a carrier wave (50 kHz, 15 Vpp) is used. By changing the amplitude of the carrier wave a constant overall change in the polarization orientation is obtained, which can be used as feedback (to lock to the zero in the differential signal) for the system.

The LC used is ZLI-2293 (a common nematic LC) doped with CB15 (a chiral nematic LC) to give a pitch of approximately 10 μm. The liquid crystal layer is 5 μm thick.

To test the system, a gas flow cell was placed at the SP active surface of the Kretschmann SP system, through which a mix of argon and nitrogen gases could be passed. The difference in RI between nitrogen and argon gas is approximately 1×10^{-5} RIU. By changing the proportions of the two gases in the mixture, the sensitivity of the method to the RI of the bounding dielectric medium can be determined. The experimental setup is shown in Fig. 1, with the polarizer after the laser oriented at 30°, as this was determined to give the greatest change in the azimuth of the ellipse through the RI change studied. The polarizer on the output of the SP system is oriented to give a zero in the differential signal corresponding to the minimum of the polarization ellipse. In the experiments presented here, the percentage of argon to nitrogen was changed in approximately 10% steps. The amplitude of the high-frequency sinusoidal voltage (feedback) needed to maintain the zero in the signal was then converted to a polarization rotation through a previously performed calibration (determined by measuring the polarization rotation caused by changing the amplitude of the high-frequency component of the applied voltage). The results of the experiment are shown in Fig. 2. The smallest measurable RI change from this data is approximately 2×10^{-7} RIU.

This sensitivity corresponds to a polarization rotation resolution of only 0.02°. This is significantly less than is possible if the SP arrangement is removed and the smallest resolvable polarization rotation is measured. The limit to the resolution is believed to be due to three main sources of noise: atmospheric changes, vibrations, and fluctuations in the gas mixture. In addition, LC director fluctuations may result in further noise; however, more work is necessary to confirm this.

The polarization rotation change as a function of the change in the RI (taken from Fig. 2) is shown in Fig. 3. According to multilayer optics modeling[6] this should be (very nearly) a straight line, so that there is clearly a discrepancy as this data shows significant curvature. It is believed that this is due to a small amount of ellipticity of the light caused by the LC cell, and that by careful control of the pitch of the LC, this ellipticity may be reduced. However, in terms of producing a chemical or biological sensor, this experiment clearly shows that even with this ellipticity, very small RI changes can still be measured.

In this letter, a liquid crystal polarization modulator has been developed for use in a differential surface plasmon ellipsometry refractive index sensor. This has overcome some problems/limitations that were previously found with the technique. A refractive index sensitivity of approximately 2×10^{-7} RIU has been achieved, with a system which is low-voltage driven, much cheaper, lighter, and more robust. In addition, the use of a liquid crystal modulator allows the possibility of multiplexing the system to allow many areas to be monitored simultaneously, leading to array sensing or imaging.

The authors would like to acknowledge the support of the Engineering and Physical Sciences Research Council.

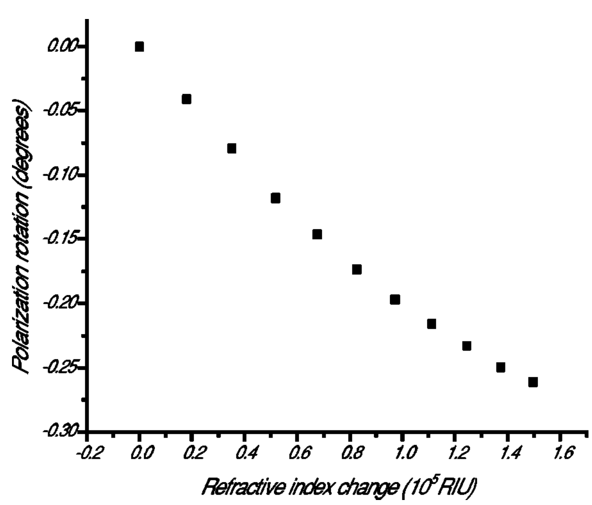

FIG. 3. The polarization rotation (obtained from Fig. 2) as a function of the refractive index of the gas mix.

This work was performed as part of a collaborative project with M. Somekh and M. Pitter of Nottingham University, UK.

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