Determination of the threshold sensitivity of a deposited dielectric film thickness control method based on surface plasmon resonance effect

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Abstract. The lower boundaries of the real time measuring range of the dielectric film thickness on the sensor surface, the principle of which is based on the effect of the excitation of surface electromagnetic waves (SEW), are determined. The simulation was performed for materials with high and low refractive index.

1. Introduction

The task of determining the thickness of thin and especially ultrathin films in real time is one of the most relevant in modern technology. Existing methods have several disadvantages. For example, the analysis of interference fringes obtained by reflecting X-rays at an almost sliding incidence, allows measuring the film thickness with an error of 1%. However, this method is applicable only for polycrystalline films no thicker than 60 nm, and the measurements strongly depend on the surface quality [1].

The X-ray diffraction method can be used to measure thicknesses with an accuracy of 0.1 nm if metal films grow with good crystal orientation. This method is very sensitive to the orientation, size of crystals and thickness changing. In addition to various X-ray methods, the radiofrequency method is widely used and based on recording quartz crystal oscillation frequency changes during film deposition. The shift of the oscillation frequency of a quartz resonator is commensurable to it mass changing. In this way, the thickness can be determined from the frequency shift if the density of the film deposited on the sensor surface and its area are known [2]. The physicochemical properties of the growing film such as phase and chemical composition, porosity have tend to change, which lead to errors in determining the thickness and offer significant disadvantage of this system.

The main disadvantage of previously described methods is that the recalculation of the obtained data into the geometric thickness of the growing film takes place. For optical coatings, optical thickness is a more important parameter. In the film growth process depending on the deposition process parameters the values of optical constants and permittivity are changing. Methods that determine the geometric thickness are not able to detect permittivity fluctuations attributable to the physical principles of measurements, which leads to a discrepancy between the properties of the obtained films and the calculated ones.

In the thickness control, there is no need for such technological processes as atomic layer deposition since the film growth process in this technology stops when the monatomic layer is reached. However, there are a large number of tasks in which the control of the deposited films
parameters is the most important. Moreover, it is necessary to observe the characteristics not only in real time, but also with very high accuracy. Consequently, it becomes necessary to create sensors, which will be sensitive to film thickness changes with an accuracy of tenths of a nanometer. The modern microelectronic industry is an area in which the implementation of this task is necessary. Non-volatile memory, photo-emitters, photo-detectors, nano-sensors, resonant-tunneling diodes, heterostructures are only a small part of the areas requiring ultra-thin films with a thickness of from several to several tens of nanometers [3].

2. Theory
Method can be used for development of fundamentally new nanoscale films thickness control systems was proposed by the authors earlier [4]. Surface electromagnetic waves (SEW) or surface polaritons are a special type of electromagnetic waves propagating along the interface surfaces. The field of such waves is localized in the near-surface layer, the dimensions of which on each side of the boundary are usually comparable to the long wavelength. Even insignificant changes in the dielectric constant affect the condition of excitation SEW due to this.

Such physical principles of the sensor operation on the SEW as sensitivity of the sensor to changes in dielectric constant make it possible to directly determine the optical thickness of the analyzed film. Using other control methods to calculate the optical thickness, additional processing of measurement results is required. An accumulation error of thickness determination occurs resulting from the impossibility of taking into account all factors leading to some changes in the physicochemical parameters of the film during it formation. These errors are especially critical for those film formation problems where high precision in tenths of a nanometer is required.

![Figure 1. Scheme of SEW excitation angle measurement experimental setup. 1 – He-Ne laser, 2 – attenuation optical filter, 3 – prism on angle rotary stage, 4 – test sample, 5 – mirror, 6 – photodetector.](image)

The experiments carried out earlier by the authors [5] confirmed the simulation results. Traditionally, a system consisting of a transparent material with a high refractive index (for example quartz) and a thin golden film deposited on its surface is used to excite the SEW. The predominant use of gold in such systems is due to the high chemical resistance of this metal.

Electron beam evaporation on the samples to excite SEW in Kretschmann geometry were applied HfO$_2$ film thickness of 1, 5 and 20 nm. The excitation angle of the surface of a clean sample, without a dielectric film on the surface of gold, was chosen as a reference point. The He-Ne laser with a wavelength of 632.8 nm was used. Measurements of the resonance angle were carried out on a setup (figure 1) with angular displacement and showed a recorded shift even for a dielectric thickness of 1 nm. Its value was 0.3 degrees and coincided with the simulation results.

The disadvantage of this scheme is the complexity of the implementation and duration of the measurement process. In order to carry out measurements in real time, an installation is necessary, which allows for one measurement we can obtain the value of the signal on the CCD matrix not for one angle, but for the whole range at once. The scheme of such an installation is shown in figure 2. The “width” of the range is determined by the aperture of the beam that illuminates the sample. One
frame obtained with a CCD array shows the dependence of the intensity on the angle of incidence and, accordingly, the position of the angle of resonance. Reading the signal several times per second allows real-time monitoring of the shift of the resonance angle.

![Diagram](image)

**Figure 2.** Scheme with aperture. 1 – He-Ne laser, 2 – telescopic system with a diaphragm, 3 – lens that focuses laser radiation on the sensor surface, 4 – sensor, 5 – image recording system (CCD matrix).

3. **Modeling**

Simulation for different wavelengths [5] showed that, with the same thickness of the dielectric on the sensor surface, the shift of the resonance angle is greater with decreasing wavelength.

![Graph](image)

**Figure 3.** The dependence of the reflection coefficient on the angle of incidence for the design of a prism K8 + 40 nm Au.

Thus, when detecting ultrathin films, it is advisable to switch to a shorter wavelength in order to increase the resolution of the system. The dielectric materials HfO$_2$ and MgF$_2$ were chosen as one of the most frequently used in the manufacture of optical coatings by sputtering. When applying 1 nm film to the sensor surface, the shift of the resonance angle is less for materials with a lower refractive index, therefore, two materials that are significantly different in this parameter were chosen for the simulation.

The task of modeling is to determine the offset of the resonance angle during the deposition of the monatomic layer. MgF$_2$ forms tetragonal crystal crystals with cell parameters $a = 0.4625$ nm, $c = 0.3052$ nm. The thickness of the monoatomic layer is assumed to be 0.3 nm. HfO$_2$ can form several crystalline modifications, but they all have approximately the same spatial measurements of the unit cell - from 0.511 nm to 0.528 nm. The value of the monatomic HfO$_2$ layer is assumed to be 0.5 nm.
Figure 4. Increasing the resonance angle when a monoatomic HfO$_2$ layer with a thickness of 0.5 nm is applied to the sensor surface. Curve 1 is the K8 + 40 nm Au prism, curve 2 is the K8 + 40 nm prism Au + 0.5 nm HfO$_2$. The plot of the curve in the region of the resonance angle is shown.

Figure 5. Increasing the resonance angle when a monoatomic MgF$_2$ layer with a thickness of 0.3 nm is applied to the sensor surface. Curve 1 is the K8 + 40 nm Au prism, curve 2 is the K8 + 40 nm prism Au + 0.3 nm MgF$_2$. Shows the plot of the curve in the region of the resonance angle.

Simulation was performed for p-polarized radiation with a wavelength of 532 nm. The material of the prism K8, the refractive index n = 1.5191. The thickness of the gold film is 40 nm, the dielectric constant $\varepsilon = -5.3 + i2.2$. The dielectric constant HfO$_2$ $\varepsilon = 4.4974$, MgF$_2$ $\varepsilon = 1.9014$. The simulation results in the WinSpall software are shown in table 1. Noticed $\alpha_0$ is the excitation angle for the SEW of a pure sensor, without a dielectric film on the gold surface, $\alpha_1$ is the excitation angle for the SEW when the monatomic layer is applied, $\Delta \theta$ is the magnitude of the resonance angle shift.

| Material   | $\alpha_0$ (deg) | $\alpha_1$ (deg) | $\Delta \theta$ (deg) |
|------------|------------------|------------------|----------------------|
| MgF$_2$ - 0.3 nm | 48.477           | 48.590           | 0.113                |
| HfO$_2$ - 0.5 nm | 48.477           | 48.875           | 0.398                |
Modern systems of registration and image processing provide high accuracy of a fixed shift of the surface plasmon resonance angle to 0.004 degrees [5]. In this way, detecting a shift of the resonance angle by 0.113 or 0.398 degrees is not a technical difficulty.

4. Conclusion
The simulation results demonstrated the high sensitivity of the proposed method for controlling the thickness of thin and ultrathin dielectric films and the possibility of confidently detecting monatomic layers of materials with fundamentally different values of the refractive index.

References
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