Possibility of measuring range extension of growing thin films thickness control, based on surface plasmon resonance effect

A E Komlev, R V Babinova and R V Dyukin
St. Petersburg Electrotechnical University “LETI”, 197376, St. Petersburg, Russia
E-mail: a.komlev@mail.ru

Abstract. Dependence of excitation angle of surface electromagnetic waves on HfO$_2$ film thickness was investigated. Modeling for different wavelengths showed the method’s applicability for real-time and high-resolution film thickness control. Proposed method can be used for development of fundamentally new nanoscale films thickness control systems.

1. Introduction
Precise control of thin films thickness is especially important task in optical devices manufacturing. For example, over a hundred of alternating layers with different refractive indexes can be used in narrow-band dielectric optical filters. Depending on operating wavelength, thickness of individual layer can varies from tens to hundreds nanometers. Moreover, thickness of layers has to be controlled with angstrom accuracy. For this purpose, two control systems are mainly used: quartz resonator and photometry. However, both these methods have some limitations. Authors have developed a method based on surface plasmon resonance, which allows calculating in real-time optical thickness of growing nanoscale film [1].

2. Theory
High sensitivity of surface electromagnetic waves (SEW) methods is based on physical principles of surface plasmons excitation [2]. Surface plasmon resonance (SPR) appears during excitation of SEW by light, which falls on layers boundaries with opposite dielectric permittivity signs. Even minor change of electro physical properties of analyzed medium causes noticeable change of excitation conditions and thus, shift of the resonance angle.

Such effect has acted as a cause to investigate the possibility of the SPR-based method applicability for thin film thickness control.

SEW was excited in Kretschmann geometry [3]. As a plasmon-generating layer, 50 nm Au film was used. Reflected from Au surface laser light intensity has a pronounced minimum, which is SPR excitation angle. During deposition of a matter on the gold surface, excitation angle increases.

As a first step, to determine the maximum thickness of HfO$_2$ film on sensor surface, at which resonance angle doesn’t exceed 80 degrees, modeling in WinSpall program was performed. Three wavelengths of laser were studied: 632.8 nm, 1064 nm and 2200 nm. HfO$_2$ was chosen as analyzed matter because of its wide utilization in dielectric optical filters from UV up to near IR regions.

For verification of the modeling results, test samples were produced and SEW excitation experiments in “glass – Au - HfO$_2$ – air” structures were performed.
3. Experiment
For the verification tests, multi-layers structures on BK7 glass were fabricated. Plasmon generating layer (Au 99.99%, 50 nm thickness) was deposited by magnetron sputtering. HfO$_2$ films with 1, 5 and 20 nm thickness were deposited on Au layer by electron beam evaporation. Thickness of HfO$_2$ films was controlled by quartz resonator system.

![Figure 1](image1.png)

**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.

Sample was fixed on prism by oil immersion, which was mounted on rotary stage with 1’ angle step. Dependencies of reflected laser light intensity on incident angle to Au surface were measured for pure Au, Au + 1 nm HfO$_2$, Au + 5 nm HfO$_2$ and Au + 20 nm HfO$_2$.

4. Modeling
Experimental results are in a good agreement with data from theoretical modeling in WinSpall software.

As a light source was chosen p-polarized He-Ne laser with wavelength 632.8 nm. Sample matter was BK7, $\varepsilon_3 = 2.2952$. Dielectric permittivity of Au, $\varepsilon_1 = -10.5 + i1.2$.

From figure 2 it is visible, that resonance angle shift, caused by deposition of 1 nm HfO$_2$ is about 0.3 degree in both theory and experiment.

![Figure 2](image2.png)

**Figure 2.** Calculated (solid curve) and experimental data comparison: 1 – sample Au/Air, 2 – sample Au/1 nm HfO$_2$/Air, 3 – sample Au/5 nm HfO$_2$/Air.
Figure 3. Calculated (solid curve) and experimental data comparison for Au/20 nm HfO$_2$/Air sample.

Existing modern ways of signal detection allows to register even smaller angle shifts [4]. Thus, experimental results confirm the possibility of detection an angstrom optical thickness changes.

Nevertheless, modeling results showed applicability limit of the method: when HfO$_2$ film thickness reaches 32 nm, resonance angle becomes over 80 degree (figure 4). Further thickness increase leads to SPR disappearance and makes detection impossible.

Figure 4. Dependence of reflectance coefficient on incidence angle to Au surface for “BK7/Au/HfO$_2$/Air” multilayer system. HfO$_2$ thicknesses are 0, 10, 20, 30, 32 nm. Laser wavelength is 632.8 nm.

The easiest available way of thickness measuring range extension is utilization of longer laser wavelength [5].

Modeling of emergence conditions of surface plasmon resonance in system glass–Au–HfO$_2$–air was also done for 1064 and 2200 nm lasers. These wavelengths exists in commonly used fiber and solid state lasers, including DPSS.

As seen in figure 5, with 1064 nm laser maximum thickness of HfO$_2$ layer rises up to 87 nm. At the same time, remains high sensitivity and possibility of precise layer thickness control.
Figure 5. Dependence of reflectance coefficient on incidence angle to Au surface for “BK7/Au/HfO$_2$/Air” multilayer system. HfO$_2$ thicknesses are 0, 10, 20, 30, 50, 70, 80, 87 nm. Laser wavelength is 1064 nm.

At laser operating wavelength of 2200 nm, thickness control limit rises up to about 220 nm (figure 6).

Figure 6. Dependence of reflectance coefficient on incidence angle to Au surface for “BK7/Au/HfO$_2$/Air” multilayer system. HfO$_2$ thicknesses are 0, 30, 60, 90, 130, 160, 190, 220 nm. Laser wavelength is 2200 nm.

Modeling results for different wavelengths and 1 nm thick HfO$_2$ film are listed below (table 1).

**Table 1.** Summarized SPR modeling results for “BK7/Au/HfO$_2$/Air” for different laser wavelength.

| Wavelength (nm) | 632.8 | 1064 | 2200 |
|-----------------|-------|------|------|
| $\theta_{res}$, thickness HfO$_2$=0nm | 43.16 | 40.95 | 41.07 |
| $\theta_{res}$, thickness HfO$_2$=1nm | 43.45 | 41.00 | 41.08 |
5. Conclusion
It was found out that increase of laser wavelength extends SPR angle shift measuring range. Thus, the maximum film thickness detection range changes from tens to hundreds of nanometers. However, with increase of wavelength, the sensitivity of the method goes down: corresponding to SPR (minimum of reflection) angle shift between 0 and 1 nm thick HfO$_2$ is 0.29º, 0.05º, 0.01º for wavelengths of 632.8 nm, 1064 nm, 2200 nm respectively.

References
[1] Komlev A E, Dyukin R V and Shutova E S 2017 *Journal of Physics: Conf. Series* **872** 012042
[2] Sharlandjiev P and Gushterova P 2003 *Vacuum* **69** 21–5
[3] Liang H Q, Liu B and Hu J F 2017 *Optik* **149** 149–54
[4] Sotnikov D V, Zherdev A V and Dzantiev 2015 *Advances in Biochemistry* **55** 391–420
[5] Nelson B P, Frutos A G, Brockman J M and Corn R M 1999 *Analytical Chemistry* **71** 3928–34