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Influence of the crystalline structure of metal films on the performance of plasmonic biosensors

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Abstract. The development of plasmonic biosensors requires careful consideration of various factors associated with the deposition of thin metal films. The quality of metal films is mostly determined by their polycrystalline structure, where smaller crystallite sizes produce higher optical losses. In present work we obtained the dependence of optical losses in thin gold and copper films on their thicknesses. These experimental results are used for the analysis of the performance of plasmonic biosensors, which demonstrate an increase in the limit of detection of up to 50% and 35% for gold and copper films, respectively.

1. Introduction

Nowadays, plasmonic biosensing has diverse applications in such areas as biochemical and pharmaceutical research, personal healthcare, veterinary, and monitoring of food quality and environment [1-3]. The performance of current plasmonic biosensors limits the range of possible applications and strongly depends on the used plasmonic materials and their optical quality. Here, we consider plasmonic biosensors based on surface plasmon resonance (SPR) excited in thin metal films by means of the Kretschmann configuration [4]. Various factors influence the optical quality of thin metal films such as growth kinetics for a particular substrate, deposition regimes, annealing, and others, because metals films with thicknesses below 100 nm deposited on dielectric substrates are mostly polycrystalline [5]. Their crystalline structure determines optical losses in metal due to electron scattering on crystallite boundaries [6-8]. Here, we study optical losses in thin gold and copper films and analyze their influence on SPR biosensing.

2. Deposition and characterization of thin metal films

Gold and copper films were deposited on silicon wafers with 285-nm-thick thermally-grown SiO_2 layer using electron-beam evaporation system Nano-Master NEE-4000. The films were deposited under vacuum conditions ($1\text{-}3\times10^{-6}$ Torr) and at a deposition rate of about 0.2 A/s. Dielectric functions of metal films were measured using spectroscopic ellipsometer VASE produced by J.A. Woollam Co in the wavelength range 300 – 1800 nm. For ellipsometry modelling, we used copper and gold dielectric functions represented by Drude Model:

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$$\varepsilon = \varepsilon_{\infty} - \frac{\omega_{\mathrm{p}}^{2}}{\omega^{2} + i\Gamma\omega}, \quad (1)$$

where the dielectric function at an infinite frequency ε_{∞}, the plasma frequency ω_{p}, and the damping rate Γ are fitting parameters. The changes in a film thickness only slightly influence the plasma frequency and the permittivity at the infinite frequency, while considerable changes in damping factors of metals respond for their varying optical properties due to the different crystalline structures of the deposited films (Figure 1) [9]. For analytical calculations, we take the plasma frequency equaling 1.55×10^{16} and 1.26×10^{16} 1/s for gold and copper films, respectively. The corresponding permittivity at the infinite frequency equals 5.5 and 5.4, respectively.

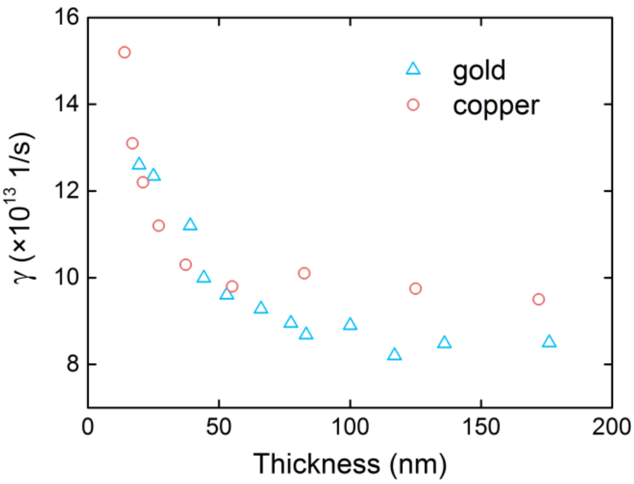

Figure 1. The damping factors for the gold and copper thin films of different thickness.

3. Plasmonic biosensing

To investigate the influence of the optical quality of metals on the performance of plasmonic biosensors, we analyse the SPR excitation in thin gold and copper films with a thickness of 47 nm. The excitation of SPR is considered according Kretschmann configuration. Using the transfer matrix model [10], we analyzed the reflection from a multilayer structure composing the following layers: 1) the glass layer with refractive index (RI) 1.523; 2) 47-nm-thick metal films; and 3) the top aqueous layer with RI of 1.33. The dielectric functions of metals are presented by the Drude model (1) with the varying damping rate in the range from 8.2×10^{13} to 12.6×10^{13} 1/s for gold films and from 9.5×10^{13} to 13.1×10^{13} 1/s for copper films. Figure 2(a) shows the SPR reflectivity curves for the gold films with different damping rates at the wavelengths of 880, 1035, 1315, and 1550 nm. The performance of SPR biosensing is characterized by such parameters as the full-width at half-maximum (FWHM) α_{FWHM} of SPR angular curve and the sensitivity to RI changes S_{RI} defined by the ratio of the shift of SPR angle $\Delta\alpha$ to the corresponding RI change of the media Δn above the metal film:

$$S_{\mathrm{RI}} = \frac{\Delta\alpha}{\Delta n}. \quad (2)$$

Both these parameters depend on the optical properties of metal films. However, variation in the damping rate results in altered optical losses in metal films determined by the α_{FWHM} while producing negligible effect on the S_{RI}. The detection limit of SPR biosensors is proportional to the figure of merit defined as the ratio of S_{RI} to α_{FWHM}. Figure 2(b) shows α_{FWHM} and FOM obtained for gold films in dependence on the damping rate of gold. For the values of Γ considered in calculations, we obtain the improvement in FOM of up to 50%. The analogous results on the SPR excitation were obtained for copper films, which have promising applications in SPR biosensing (Figure 3) [11-12]. For the considered damping rates of copper films, the FOM can be improved up to 35%.

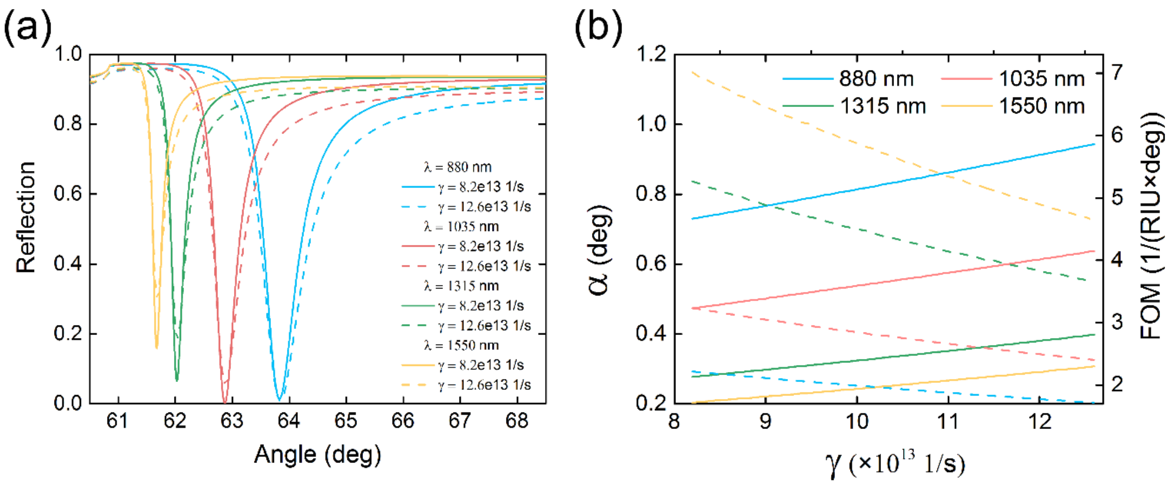

Figure 2. (a) SPR angular reflectivity curves for gold films characterized by different damping factors and at different wavelengths. (b) Full-width at half-maximum of SPR reflectivity peaks and figure of merit for SPR biosensing based on thin gold films characterized by different damping factors.

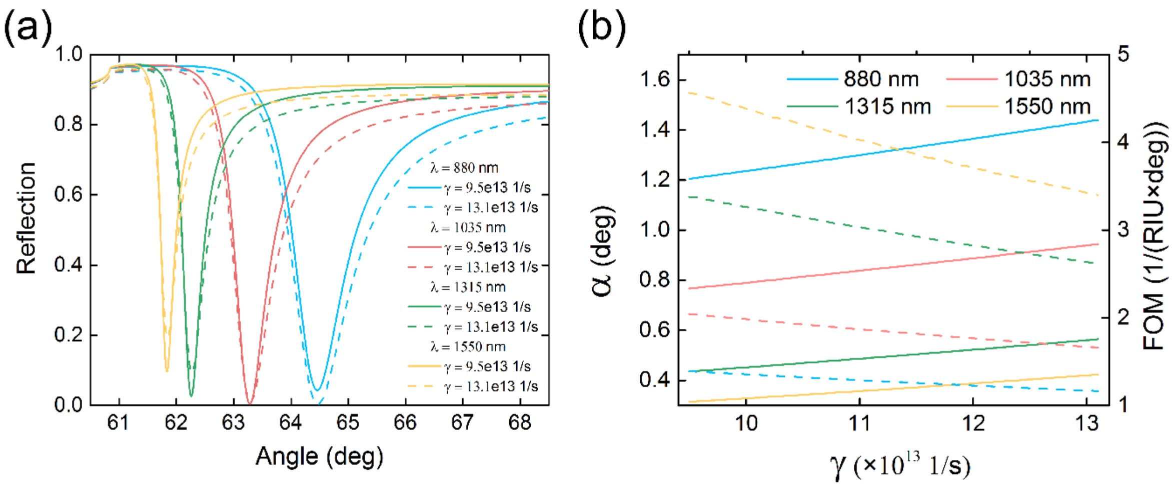

Figure 3. (a) SPR angular reflectivity curves for copper films characterized by different damping factors and at different wavelengths. (b) Full-width at half-maximum of SPR reflectivity peaks and figure of merit for SPR biosensing based on thin copper films characterized by different damping factors.

4. Conclusions

The performance of plasmonic biosensors depends on the optical quality of metal structures used for SPR excitation. In the case of thin metal films, a number of factor determines their optical properties, which is mostly reflected in their crystalline structure formed during the deposition process. In present study, we used the Drude model for the description of dielectric functions of gold and copper. The variances in damping rates for gold and copper films were obtained using ellipsometric measurements. Thereafter, using the obtained optical parameters, we analyzed the SPR excitation in thin metal films with application to optical biosensing. For the experimentally obtained variances in the damping rate of gold and copper, the difference in the FOM characterizing biosensing efficiency can reach up to 50% and 35%, respectively. Due to this, the development of plasmonic biosensors as well as other plasmonic and optoelectronic devices based on thin metal films requires the optimization of film deposition process in order to reach the best performance.

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