Vacuum 138 (2017) 55–63

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Confocal sputtering of (111) orientated smooth gold films for surface plasmon resonance approaches

G. Schmidl [a, *], J. Dellith [a], N. Teller [a], A. Bochmann [c], S. Teichert [c], O. Stranik [a], A. Dathe [a], V. Tympel [b], F. Schmidl [b], E. Kessler [a], T. Wieduwilt [a], W. Fritzsche [a]

[a] *Leibniz Institute of Photonic Technology (IPHT), Albert-Einstein-Straße 9, 07745 Jena, Germany*
[b] *Ernst Abbe Hochschule, University of Applied Sciences, Carl-Zeiss-Promenade 2, 07745 Jena, Germany*
[c] *Friedrich Schiller University of Jena, Institute of Solid State Physics, Helmholtzweg 5, 07743 Jena, Germany*

ARTICLE INFO

Article history:
Received 1 November 2016
Received in revised form
23 January 2017
Accepted 24 January 2017
Available online 25 January 2017

Keywords:
Gold films
Crystalline texture
Resistivity
Annealing
Confocal sputtering
Dielectric function

ABSTRACT

We report on the influence of sputtering parameters on the microstructural growth of 200 nm thick Au layers and, moreover, on their crystalline, electrical, and optical properties using a confocal sputtering arrangement. The confocal sputtering arrangement allows the production of highly homogeneous layer thicknesses that depend on pressure, power, and target-substrate distance. Layers deposited at low Ar pressure show extremely smooth and densely-packed films, as well as a preferred {111}-texture contrary to films deposited at higher Ar pressure. Furthermore, an increase in electrical resistivity combined with a decrease in grain size is observed for these layers. A subsequent annealing process up to a temperature of 600 °C reduces the resistivity, increases the grain size, improves the fraction of {111}-texture, and significantly changes the surface morphology for all initial states. Investigations at 4.2 K showed that the decrease in resistivity is caused by a structural change that takes the number of grain boundaries into consideration. The dielectric function investigated by ellipsometric measurements is dependent on the deposition process parameters, especially on Ar pressure. Surface plasmon resonance calculations based on the layers produced using the Otto configuration show the best performance using a specific setup for layers deposited at low Ar pressure.

1. Introduction

Several technologies of molecular sensing in the life sciences or environmental safety exist that use the propagation of photon-electron waves (surface plasmon polaritons - SPPs) along a metallic interface [1–3]. A crucial factor in SPP propagation and these sensing applications is the flatness of the metal layer surface. The degree of flatness affects the propagation length of the plasmon, and therefore, the sensing efficiency. It is advantageous to minimize any damping effects on its propagation path. Nevertheless, precise structured surfaces can be used in specific surface plasmon resonance (SPR) approaches, for instance to manipulate SPPs, but also in the Surface Enhanced Raman Spectroscopy (SERS), for field enhancement, or in the Localize Surface Plasmon Resonance spectroscopy (LSPR) [4,5]. In general, surface inhomogeneity should be reduced in order to avoid surface scattering and facilitate better light coupling efficiency and wave propagation. The advantage of smooth surfaces and additionally a high crystallinity leads to higher conductivity and lower energy losses. This is essential for SPP, as well as for patterning by etching or FIB milling [6,7]. Gold is one of the few metals that is relatively easy to produce in epitaxial form or with a preferred crystalline orientation. In addition, it is chemically inert, compared to silver, and is the material of choice for plasmonic devices, because of its special affinity to biomolecules. Consequently, gold is often used in bioanalytics.

The influence of the surface morphology of Au layers and the layer properties in general on the optical response can be described by analyzing the dielectric function [8]. The propagation length of the SPP wave ($\delta_{SPP} \sim \varepsilon_1^2/(2{\cdot}\pi{\cdot}\nu{\cdot}\varepsilon_2)$) is expressed via the incident frequency (ν) and both the real part (ε_1) and the imaginary part (ε_2) of the dielectric function of the layer material. It is therefore, significantly influenced by the layer quality, in other words by the material properties which are themselves influenced by the deposition parameters [9–11]. In order to compare the performance of different materials in plasmonic devices, the quality

* Corresponding author.
E-mail address: gabriele.schmidl@leibniz-ipht.de (G. Schmidl).

http://dx.doi.org/10.1016/j.vacuum.2017.01.025
0042-207X/

factors Q_{SPP} of the layers, or figures-of-merit, can be used. Q_{SPP} is proportional to the propagation length and thus is also characterized by ε_1 and ε_2 [12,13]. It is given by

$$Q_{SPP} = {\varepsilon_1}^2(\lambda)/\varepsilon_2(\lambda) \sim \delta_{SPP}. \quad (1)$$

The absorbance of metals and consequently the resulting optical losses, to which the Drude model for conductivity is applied, can be influenced by surface scattering and other film defects. Such conductivity modeling is described for example in Refs. [14–17]. Statements about loss mechanisms (crystal defects or surface scattering) are evident in x-ray diffraction measurements, as well as in resistivity measurements, especially, those that are temperature dependent. It is also known that the layer growth is influenced by different deposition parameters such as temperature, pressure, power, and throw distance (Thornton model). Post annealing is a further process which may be used to affect the layer properties [18–20].

In this paper, we present the results of investigations into the crystalline properties, surface morphology, and electrical properties of Au films, as well as their optimization via an annealing process. The objective is to find a parameter set that produces films with a preferred crystal orientation and with very smooth surfaces using a confocal magnetron sputtering technique. The investigation of the optical constants should show the influence of preparation conditions on the quality factor of gold films for plasmonic devices.

2. Materials and methods

2.1. Angled sputtering of Au films

A load-lock magnetron sputtering system (VON ARDENNE Anlagentechnik GmbH) was used for layer deposition. Compared to rating scales of other manufacturers the built-in magnetrons (PPS-A 100) are of the "extremly balanced" type. In general, the confocal sputtering technique used enables the deposition of films made of different materials all at once or by co-sputtering to generate complex film systems in one step. Furthermore, the target area does not need to be larger than the substrate in contrast to conventional top-down sputtering. In our equipment the targets are arranged at a fixed angle (θ_T: 30°) to the substrate normal. Thus, for this angled sputtering it is necessary to offset the tilted magnetron from the center of the rotational axis and to direct it to a spot (approximately 0.5× substrate radius) on a rotating substrate. The angle θ_T is optimized in order to deliver a highly uniform film thickness, and thus, in our case the uniformity can only be influenced by the target-substrate distance. To simplify these conditions, an optimized, constant target-substrate distance (TSD or throw distance) of 85 mm was investigated and used for all experiments.

A cryogenic pump system generates a background pressure of less than $1\cdot10^{-5}$ Pa in the process chamber. System parameters such as Ar pressure (p_{Ar}) and sputtering power (P) were varied to investigate the layer properties. The Ar pressure was varied in the range from 0.5 Pa to 5 Pa and the power in between 100 W and 500 W (1.3–6.4 W/cm^2). The film thickness used was fixed at 200 ± 10 nm. On that basis further comparisons can be better performed.

Since the substrate carriers, and hence the substrates themselves, were passively cooled by cooling the rotating table, the deposition conditions do not influence a later annealing process. Si wafers that are (001) orientated and glasses were used as substrate materials. In any case in order to prevent short-circuits during four-point resistivity measurements the Si wafers were thermally-oxidized (thickness of 1 μm).

2.2. Film characterization methods

The investigation of film structure and crystalline film properties was supported by a number of complementary methods. Surface sensitive techniques, above all scanning force microscopy (AFM), were used for morphological evaluations including quantitative results concerning film surface roughness. An AutoProbe CP system from Park Scientific operating in contact mode with sharpened micro lever tips (tip radius below 10 nm) was used. SEM measurements via field-emission scanning electron microscopy (FE-SEM; JEOL JSM-6300F) were used for both the investigation of the film topography and the growth mechanism. To achieve this, cross-sectional samples were taken after short-term LN_2 cooling in order to reduce the degree of gold ductility and create true fracture patterns. The crystalline structure was investigated by X-ray diffraction (XRD; Panalytical X'Pert Pro for crystallite size estimation; Bruker D8 advance for texture analysis) with Cu-K$\alpha_{1,2}$ (Kα_1: 1.5406 Å) radiation. The Scherrer equation was used for the estimation of the crystallite sizes. A quantitative estimation of the volume fraction of the fiber texture was done using the component fitting method [21] utilizing the software MULTEX from Bruker.

2.3. Annealing process and resistivity measurements

The samples were annealed at 200 °C, 400 °C, and 600 °C in a nitrogen environment to investigate the influence of a change in microstructure on electrical resistance behavior via annealing. Each annealing step took 1 h and the resistivity ρ was determined after each annealing process by a four-point setup at room temperature (RT). The temperature-dependent resistance measurements were performed by a cryogenic measurement setup from 4.2 K to RT.

2.4. Optical measurements

The as-deposited Au films were investigated via ellipsometry (SE850, SENTECH) in order to determine the optical constants and thus the optical properties depending on the processing conditions. The refractive index n and the absorption coefficient k were measured, and the dielectric function

$$\varepsilon_m(\lambda) = \varepsilon_1(\lambda) + i\varepsilon_2(\lambda) \text{ with} \quad (2)$$

$$\varepsilon_1(\lambda) = n^2(\lambda) - k^2(\lambda) \text{ and} \quad (3)$$

$$\varepsilon_2(\lambda) = 2\cdot n(\lambda)\cdot k(\lambda) \quad (4)$$

[4,22] was calculated. Here, ε_1 describes how strongly the material is polarized, and ε_2 describes the losses in the material. For calculations and for statements about general properties of surface plasmon resonances (SPR) the investigated gold layers were used. Due to the layer thicknesses, therefore the Otto configuration was selected. In this configuration, polarized light is coupled via a prism and a small air gap into a surface of the thick gold layer in order to characterize the system with respect to the SPR efficiency.

In this case, the SPR is dependent on the angle of incidence and strongly dependent on the gap and the media between the prism and Au layer, in which the biomolecules can be introduced to be examined. The range of dimensions of this dielectric gap using our Au layers is between a few hundred nm and 1 μm for SPR generation. In general, the reflection of spectrally broad light from the system has a minimum at a particular wavelength since the radiated light is absorbed and converted to density fluctuations occurring near the layer surface.