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Research paper

Towards sub-micrometer high aspect ratio X-ray gratings by atomic layer deposition of iridium

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ABSTRACT

X-ray grating interferometry is an excellent technique for X-ray phase contrast imaging and X-ray wavefront sensing with applications in materials science, biology and medical diagnosis. Among other requirements, the method depends on the availability of highly X-ray absorbing metallic gratings. Here, we report on the fabrication and characterization of high aspect ratio iridium gratings with a period of 1 μm and a depth of 30 μm combining deep reactive ion etching of silicon and atomic layer deposition of iridium. The implementation of such structures can greatly enhance the sensitivity of grating-based X-ray phase contrast imaging and thus, expand further its broad range of applications.

1. Introduction

X-ray grating interferometry [1–3] is a prominent method for X-ray wavefront sensing [4,5] and X-ray phase contrast imaging with applications in materials science [6], biology and medical diagnosis [7–9]. The technique crucially depends on the availability of highly X-ray absorbing metallic gratings. For absorbing typical X-ray photon energies of 20 keV and above, thicknesses of at least 25–30 μm of a high atomic number element, such as gold, are required. On the other hand, typical X-ray grating periods range from 15 down to a few micrometers, and thus, high aspect ratio structures are necessary for most grating-based X-ray phase contrast imaging setups. Recently, X-ray phase contrast imaging applications using even smaller grating periods have been proposed [8,10] but current fabrication methods cannot routinely produce gratings with periods of 1 μm and below. To date, X-ray absorption gratings are commonly fabricated combining optical or X-ray lithography with gold electroplating in silicon templates [11,12] or polymer resist molds [13]. However, such fabrication techniques are not easily transferred for the production of sub-micrometer high aspect ratio structures. Here, we propose and demonstrate the production of high aspect ratio (>50) iridium gratings with a period of 1 μm and a depth of 30 μm combining deep reactive ion etching (RIE) of silicon and atomic layer deposition (ALD) of iridium [14]. Until now, ALD processes have been successfully applied to conformally deposit oxide layers on high aspect ratio (>180) nanostructures [15,16] and ALD metal coatings have also been proven very effective to fabricate high resolution diffractive X-ray optics in the sub-100 nm structure size range [17,18]. In this work, we thoroughly optimized the ALD recipe to extend the conformal metal coating to trench depths of tens of micrometers while keeping the 1 μm grating periodicity. After scanning electron microscopy inspection, the fabricated gratings were successfully implemented and characterized in a laboratory X-ray phase contrast imaging setup.

2. Materials and experimental methods

2.1. Grating-based phase contrast X-ray imaging

The interaction of X-rays with an object can be described by introducing a complex refractive index, $n = 1 - \delta + i\beta$, in which β and δ respectively account for the absorption and phase shift undergone by the X-ray wavefield due to the specimen [19]. For biological soft tissues with small density differences, the phase shift refractive index variation δ can be up to three orders of magnitude larger than its absorption counterpart β, thus allowing X-ray phase contrast

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imaging to deliver images with much higher contrast in comparison to those obtained by conventional X-ray absorption imaging. Among different existing X-ray phase contrast imaging techniques, X-ray grating interferometry yields excellent contrast and quantitative information of the sample under investigation [20,21]. Grating-based phase contrast imaging can be both performed in synchrotron radiation facilities [2] and using incoherent laboratory X-ray sources [22]. In the latter case, the setup typically requires the use of two highly absorbing X-ray gratings, G_0 and G_2, and a phase-shifting grating, G_1, as schematically represented in Fig. 1. The combination of G_0 and G_1 gratings creates an X-ray intensity modulation at a given downstream distance (Talbot distance) from the G_1 phase-shifting grating. The G_2 grating is necessary to detect the intensity modulation when the pixel size of the X-ray detector is larger than half the period of this X-ray intensity modulation. After inserting the sample near the G_1 phase-shifting grating, the X-ray intensity modulation is modified. This change allows the detection of the sample differential phase contrast image by step-scanning one of the three gratings in small steps covering a range of one or several grating periods while recording consecutively the transmission X-ray intensity images [2]. The exact geometry and grating periods requirements for an X-ray grating interferometer setup can be calculated using the formulas reported elsewhere [23].

The fabricated X-ray gratings were tested in a laboratory setup using a Hamamatsu L10101 X-ray microsource with a tungsten target and an expected focused source size of 5–10 μm. The X-ray microsource high voltage was set to 40 kV and an electron current of 0.200 mA was employed. The X-ray images were acquired using a scintillator-based sCMOS camera with pixel size of 22 μm (1024 × 1024 pixels) from Photonic Science Ltd. (UK). The gratings were mounted on motorized stages with 20 nm positioning accuracy for alignment and scanning simplicity. The gratings periods p_0, p_1 and p_2 were chosen to be 1 μm. Thus, two absorbing gratings made of iridium were mounted in our laboratory X-ray phase contrast imaging setup as G_0 and G_2. The phase-shifting G_1 grating was made of silicon and it had a thickness of 25 μm, thus producing the required phase shift of π rad for an X-ray energy of approximately 20 keV.

2.2. Fabrication of the X-ray gratings

Phase contrast imaging X-ray gratings require thin supporting substrates, made of silicon, to minimize the X-ray absorption when typical photon energies below 30 keV are employed. Nevertheless, the handling of very thin substrates can be challenging because of their fragility and finally, 4-inch silicon double side polished wafers with a thickness of 250 μm were used to fabricate the X-ray gratings reported here. The schematics of the fabrication process is depicted in Fig. 2. Before the lithography step, the silicon wafers were coated with a 100 nm thick layer of chromium by electron beam evaporation. After that, the grating pattern was produced using conventional UV photolithography on positive photoresist layer (MicroChem Corp. S1805) using a Karl Suss Mask Aligner MA6 in vacuum contact mode. After exposure and development of the photoresist, the pattern was further transferred into the chromium hard mask by a RIE Cl_2/CO_2 based process. Then, the photoresist residuals were removed by immersion in acetone and isopropyl alcohol. Using the patterned chromium layer as hard mask, the high aspect ratio silicon structures were produced by a SF_6/C_4F_8 based deep RIE, also commonly known as Bosch process, using an inductively coupled plasma (ICP) Plasmalab 100 system from Oxford Instruments Plasma Technology (UK). The high aspect ratio and high resolution structures were achieved after a careful fine tuning of all process parameters such as the gas flows, chamber pressure and radiofrequency powers [24]. As schematically suggested in Fig. 2 (c), the photolithography and deep silicon RIE processes were adjusted to produce a 2 μm period silicon grating with a duty cycle of 0.25. In this way, the 1 μm iridium grating was only produced after a conformal metal coating at both sides of the silicon lines. Since silicon is a very low absorbing material in comparison to iridium, the gaps and the silicon lines of the grating can be regarded as equivalent for the X-ray wavefield that will only be sensitive to effective iridium grating period. In the past, such an approach has been successfully demonstrated for the production of high resolution X-ray diffractive optics [17,18]. Finally, the last step in the grating fabrication was the conformal deposition of iridium by ALD using iridium acetylacetonate, $Ir(acac)_3$, and oxygen gas, O_2, as precursors [14], as depicted in Fig. 2 (d). In principle, this deposition method can be used to coat conformally any surface structure by repeatedly supplying two complementary reactant vapors in alternating pulses. Because the chemical reactions are forced to happen entirely on the surface and they are self-limited by the amount of precursor reactant that can be adsorb by the surface, the technique is well-suited for a film growth with almost atomic monolayer accuracy [25,26]. During the investigations presented here, a Picosun™ R-200 Advanced ALD tool with the capability of delivering a plasma-enhanced oxygen precursor was used. This ALD system has a large reaction chamber suitable for up to 8-inch wafers and it has 6 separate inlets allowing for several precursors to be supplied. The inlets are constantly flushed with a flow of nitrogen, even during the ALD cycle steps, to prevent the growth of materials and the likely clogging of the precursor inlet orifices. As a result, the reactor chamber also requires constant vacuum pumping to prevent an excessive pressure increase, which is kept at around 10 mTorr. Since the precursor species are supplied by mixing the precursor vapor with inlet flow of nitrogen, the constant vacuum pumping of the reactor chamber is artificially limiting the precursor exposure of the surface being coated. However, the system can be operated using a Picoflow™ diffusion enhancer operation mode which consists of: (1) the temporary closure of the vaccuum pumping valve during the precursor delivery pulse of every ALD cycle; and (2) the addition of a stainless steel lid to reduce the reactor chamber size. Using this operation mode, the effective dosing time is increased and the diffusion of the precursor species is promoted. Concerning the precursor species, the $Ir(acac)_3$, that is a powder at room temperature, is kept at high temperature (195 °C) and low pressure, so that it sublimates inside its reservoir. The precursor is delivered by shortly opening the container valves and mixing the $Ir(acac)_3$ vapor with nitrogen flow through the line. Therefore, the $Ir(acac)_3$ vapor pressure limits the total amount of precursor that can be delivered for a single ALD pulse to the reactor chamber. The O_2 precursor can be both delivered as gas for an exclusively thermal reaction or through a plasma generator for a

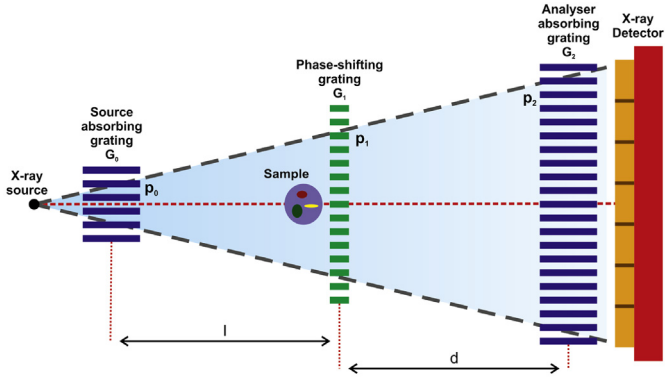

Fig. 1. Schematics of laboratory grating-based phase contrast X-ray imaging setup composed of two highly absorbing X-ray gratings, G_0 and G_2, and a phase-shifting X-ray grating, G_1 (drawing not to scale).