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Fabrication of narrow-gap nanostructures using electron-beam induced deposition etch masks

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ABSTRACT

We present an optimized procedure to create narrow-gap nanostructures using electron-beam induced deposition etch masks. Incorporation of a Cr sacrificial layer in between Au metal layer and EBID mask improves the etch mask lithography in terms of dimension control and surface flatness. EBID mask writing is performed with a single-pass strategy varying the dose during writing. This patterning strategy allows us to create relatively thick EBID masks with steep, narrow gaps. An argon sputter etch is used to transfer the mask into the underlying material where the low etching rate of sacrificial layer compared to the EBID mask improves transfer of the mask pattern into the metal nanostructure. A Cr wet etch step is then used to remove sacrificial layer and EBID mask material. We show fabrication of Au dimer nanostructures with gap spacing well below 10 nm, down to about 5 nm, on Si substrates as well as ITO-coated glass. This method may enable the fabrication of nanostructures with small gap spacing and high degree of material purity and flatness on arbitrarily shaped surfaces.

1. Introduction

Electron-beam induced deposition (EBID) is a direct-write nanofabrication technique that can reproducibly reach resolution below 10 nm. With EBID, a focused electron beam decomposes precursor molecules adsorbed on a substrate, leaving a patterned nanostructure as a deposit. Comprehensive reviews of EBID can be found in van Dorp et al. [1], Silvis-Cividjian et al. [2] and Utke et al. [3]. EBID resolution of ~1 nm has been reached on a free-standing thin film [4], and 3 nm on a solid substrate [5]. Also fabrication of 3 nm lines and spaces has been demonstrated [6]. In addition, EBID is a 3-dimensional patterning technique and can be carried out on complex shaped, non-planar substrates [7]. However, a major issue in EBID is to control the purity of the deposit, which typically contains a large fraction of carbonaceous material resulting from uncontrolled decomposition of the precursor (usually organo-metallic molecules). Especially for the important class of metal (plasmonic) nanostructures, purity is important to achieve high conductivity and avoid damping of the plasmon resonance. Precursor molecules reach maximum ~95 at.% Au directly after adsorption [8,9,10]. Post-deposition annealing can improve this factor, but this comes at the expense of size control [11–14]. Also, excess material may remain on the substrate at the end faces of the nanostructures as noticed by Graells et al. and the extinction coefficient at optical frequencies might still be higher compared to crystalline Au and evaporated Au layers.

Electron-beam-induced deposition mask lithography (IDML) has been suggested as an alternative to direct-write EBID of pure, single component, nanostructures [15,16,17]. In IDML, EBID is used to define a high-resolution mask on a native metal film and this mask is subsequently transferred to the metal film using plasma etching techniques.

Using IDML, Au bowties on AFM tips and cone shaped structures on non-planar substrates have been demonstrated [16,18]. However, so far sub-10 nm gap spacing using IDML has not yet been achieved. Pattern transfer depends on the mask material and mask removal may be an issue. Sensitivity to proximity effects in the mask layer may give rise to irregularities in the nanostructure height, i.e. a rough upper surface.

Other techniques like EBL can be used to create structures with small dimensions and spacing, however EBL is difficult to use on non-flat substrates [19]. FIB milling is an alternative, but gallium contamination of the structure might be an issue and spacing or gap sizes are limited to about 10 nm [20,21]. Nevertheless, nanostructures with small gaps and spacing are attractive for applications like optical nano-antennas, because the electric field in the gap will be enhanced. High electric field enhancement and thus small gaps are desirable for applications such as photo detection, light emission and light harvesting [22].

A crucial aspect in both study and application of nanodevices like optical antennas is the availability of a fabrication technique that can reproducibly meet three criteria. First, for spectral tuning in the visible, antenna dimensions should be controlled in the 10 nm size range [23]. Second, for optimal performance, i.e., maximum electric field enhancement, dimensional control even down to sub 1 nm gap sizes is required [24]. Third, control over the morphology, surface roughness and material quality is important to have a good quality factor resonator [25,26,27].

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In order to meet these three requirements, we propose the use of an additional sacrificial layer between the EBID etch mask and the metal layer in the IDML process. In this way, we achieve improved spatial control over nanostructure dimensions including small gap spacing. Below, we describe the fabrication procedure and the results of this fabrication process.

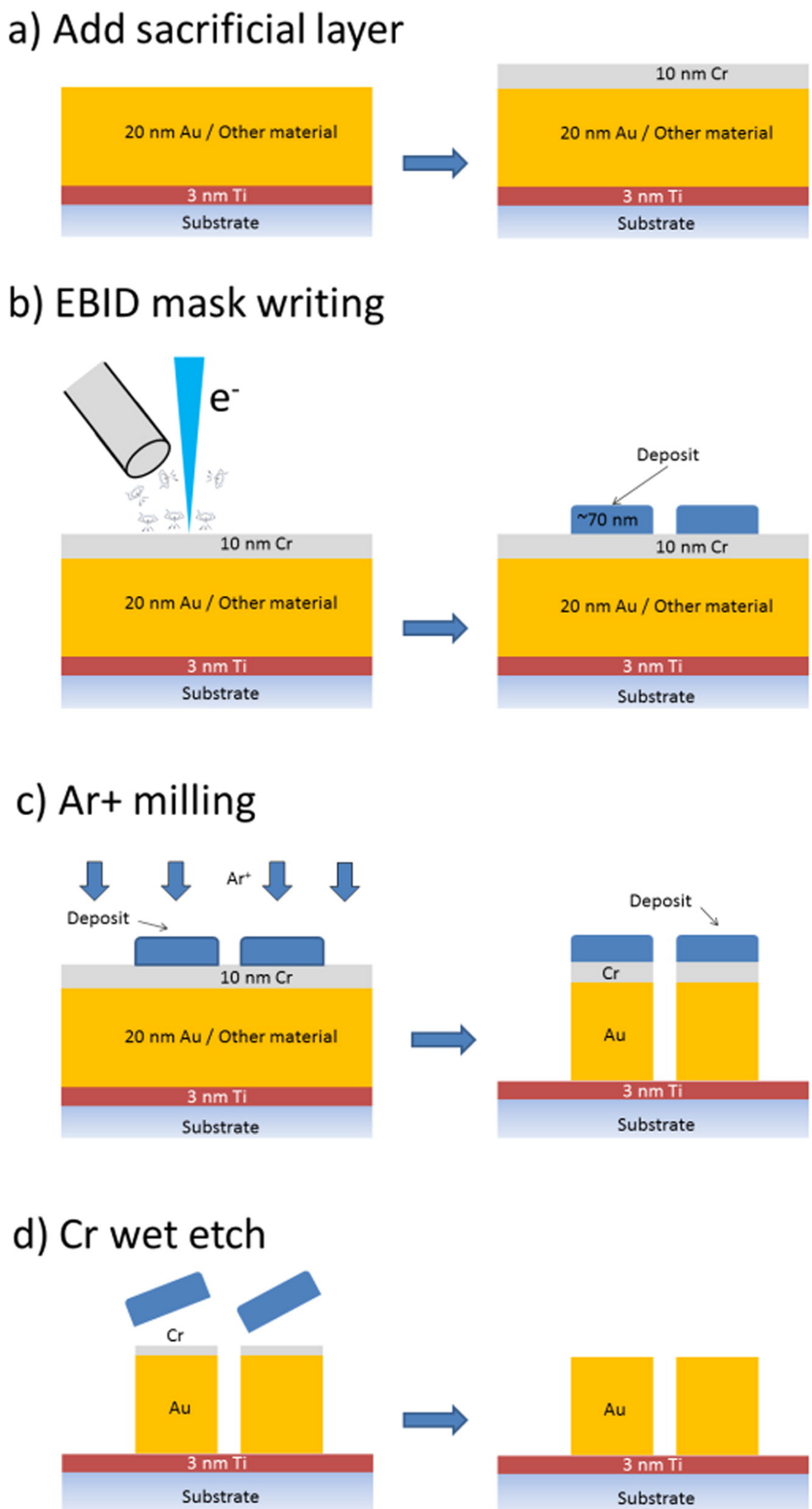

Fig. 1. Sketch of the steps in the fabrication method. (a) A sample consisting of a 10 nm chromium sacrificial layer on top of a metal in which we want to fabricate a nanostructure. (b) The EBID mask is written on the sample using a $MePtCpMe_3$ precursor gas the mask consists of two lines with a gap. Note that layer thicknesses are not to scale, but typical dimensions are indicated. (c) An argon sputter etch transfers the mask in the underlying layer. (d) A wet etch is used to remove the sacrificial layer and lift-off the remaining EBID material.