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Fabrication of Sub-20nm Metal Nanogaps from Nanoconnections by the Extended Proximity Effect*

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Abstract: We describe the fabrication of metal nanogaps of sub-20nm in feature size using the proximity effect in electron beam lithography (EBL). The proximity effect is extended to develop a flexible and practical method for preparing metal (e.g. Au or Ag) nanogaps and arrays in combination with a transfer process (e.g., deposition/lift-off). Different from the direct gap-writing process, the nanogap precursor structures (nanoconnections) were designed by GDSII software and then written by electron beam. Following a deposition and lift-off process, the metal nanogaps were obtained and the nanogap size can be lowered to ~10nm by controlling the exposure dose in EBL.

Key words: metal nanogap; nanofabrication; proximity effect; electron beam lithography
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1 Introduction

Recently, nanostructures, nanomaterials, and nanofabrications have been developed and have attracted considerable attention in the fields of nanotechnology and device application[1~5]. The development of molecular electronics and single electron devices[6,7] has been a challenge for the single nanostructures[8] and even single molecules[9,10]. The nanogaps have provided an important platform for investigating the physical properties of individual functional molecules and nanostructures (e.g., quantum dots, nanotubes, and nanowires)[11~13]. At the same time, the fabrication of the nanogaps and their alignments are essential to develop quantum devices from individual nanostructures or single molecules. Lithography techniques involving soft lithography and nanoimprints have been successfully used to prepare nanogaps. Electron-beam lithography (EBL)[12,14] is an important and flexible technique for fabricating nanogaps with high resolution. Moreover, the EBL process can overcome its low writing speed and high cost in a large area by use of a subsequent nanoimprint process. However, it is still difficult to prepare narrower nanogaps (e.g., sub-20nm and even sub-10nm) by the direct writing procedures during the EBL process.

In this paper, we describe an alternative method, involving EBL and a lift-off process, to fabricate metal nanogaps of sub-20nm, and even down to ~10nm in size. Here, the proximity effect[15] in the EBL technique is extended and used to achieve the precursor nanoconnections, and thus metal nanogaps followed by a deposition and lift-off process[16,17]. In addition, the size of the nanogaps could be lowered to ~10nm by the combination of the structural design and the EBL development processes. We expect that the as-prepared metal nanogaps will bring some alternative opportunities in the studies on single molecules and single electron nanodevices.

2 Experiment

Metal nanogaps were fabricated by a standard EBL process followed by a deposition and lift-off procedure. In all EBL cases, we used FEI Sirion scanning electronic microscopy (SEM) with a Raith Elphy quantum pattern generator for the preparation of the nanogap precursor structures (nanoconnections). The precursor nanoconnection was designed to rely on the proximity effect, as shown in Fig. 1. A typical fabrication schematic process is shown in Fig. 2. A bilayer-

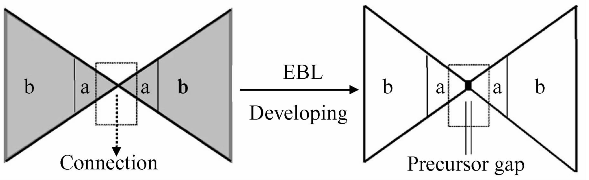

Fig. 1　Schematic outline of nanogap precursor structures derived from the designed connection between two features due to the proximity effect in EBL

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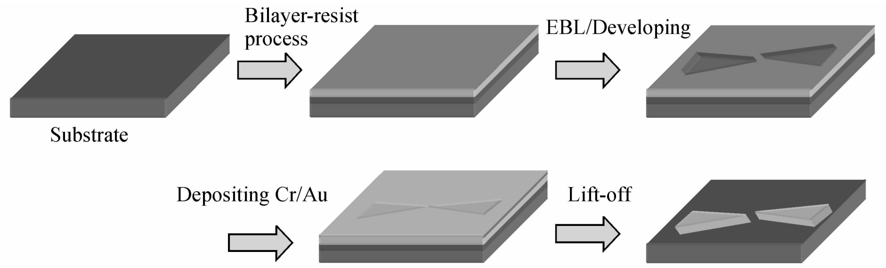

Fig.2 Schematic diagram of the fabrication of nanogaps by the proximity effect in EBL

resist process was used to coat one bottom layer of poly (methyl methacrylate-methacrylate acid) (PMMA-MA) and one top layer of poly (methyl methacrylate) (PMMA, Mw: 430K) on the silicon wafer. The whole layer thickness of the resist was about 300nm. Then, the EB writing was carried out at an accelerating voltage of 30kV and a beam current of 160pA. We varied the nanoconnection or gap size by varying the writing dose in the range of 100 ~ 1000μC/cm^2. After the e-beam exposure, all samples were developed in a mixture of methyl-iso-butyl-ketone (MIBK) and isopropanol (IPA) (volume ratio 1 ∶ 3) for 1min at room temperature, followed by washing in IPA for 20s. Finally, metallic nanogaps were obtained by a lift-off process after the sputtering deposition of metal (Cr/Au, thicknesses 5/30nm). An FEI Sirion200 SEM was used to investigate the morphologies of the resulting nanostructures.

3 Results and discussion

In a typical EBL process, the size of the developed nanostructures become smaller than that of the precursor nanoconnections designed by GDSII software due to the proximity effect. In the fabrication of the uniform nanostructures using EBL, one usually tries to avoid the proximity effect. However, herein, we used the proximity effect to fabricate nanoconnections for the nanogaps and their arrays. Figure 1 and Figure 2 show the scheme of the formation of nanogap precursor structures by the proximity effect in EBL. Different from the process in other reports[15], we designed the connection between the two triangular pads, not the direct gap nanostructures. The exposure dose in part (a) was larger than that for part (b). Part (a) should be under exposure when we expose the designed structures in the same dose. For the two connected triangular pads, the originally connected parts in the dashed field in Fig. 1 could be changed in the developed PMMA cases when the dose was exact for part (b). In other words, the size of the central part will be controlled for the fabrication of nanogap precursor structures when varying the writing dose.

Figures 3 (a) and 3(b) show the typical SEM images of the precursor nanoconnections and their ar-

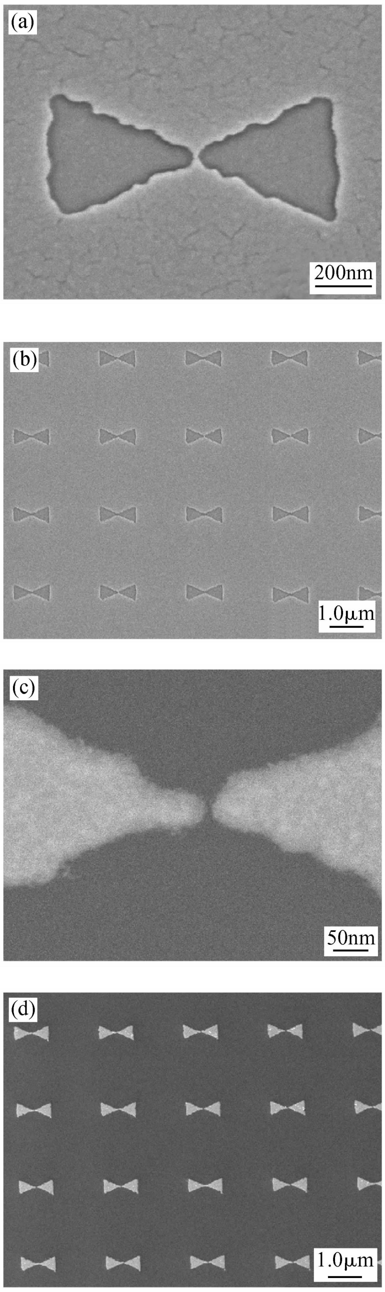

Fig.3 (a) SEM image of the nanoconnection; (b) SEM image of the array developed by EBL at a writing dose of 700μC · cm^{-2}; (c) Cr/Au nanogap; (d) Its array fabricated after the metal deposition and lift-off process

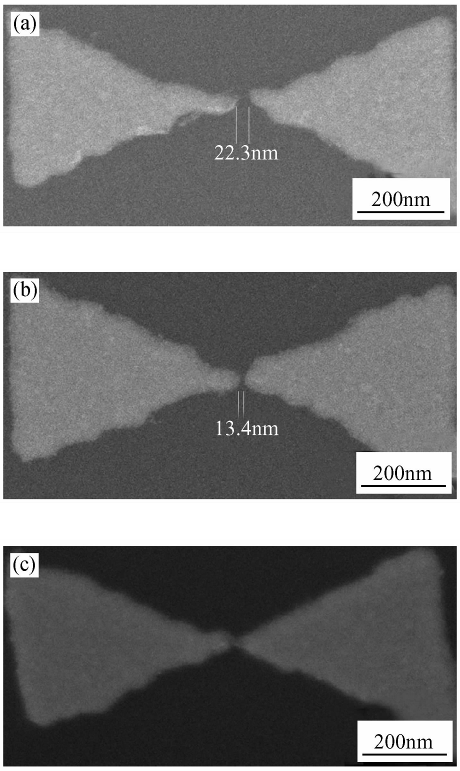

Fig. 4 SEM images of metal nanogaps by varying the writing dose followed by a deposition/lift-off procedure (a) 400μC $\cdot$ cm^{-2}; (b) 500μC $\cdot$ cm^{-2}; (c) 1000μC $\cdot$ cm^{-2}

ray before the formation of the metal nanogaps. Relying on the proximity effect in EBL, a writing dose of 700μC/cm^2 was used to obtain the nanogap precursor structures of smaller than 20nm in width between the two triangular pads after they were developed in the mixture of MIBK and IPA. We transferred the positive precursor structures into a nanogap by metal sputtering (Cr/Au with 5/30nm thick) and a lift-off process. Figure 3 (c) presents the metal nanogap of 12nm obtained after the lift-off process and Figure 3 (d) shows a typical array of the resulting metal nanogaps.

As mentioned above, the sizes of the metal nanogaps can vary in a wide range by adjusting the writing dose and the designed structures. This is an important advantage in our fabrication process. Moreover, we do not need to design the original nanogaps to achieve the nanogaps with a gap size in the range of 10～50nm. Figure 4 shows some typical SEM images of different metal nanogaps based on the proximity effect of EBL and the deposition/lift-off procedure. When we increased the writing dose from 100 to 700μC/cm^2, the size of the resulting metal nanogaps decreased from 50 to 10nm, as illustrated in Figs. 4 (a) and 4(b). When increasing the writing dose to 1000μC/cm^2, however, we only obtain a nanoconnection between two metal triangular pads due to a complete exposure in part (a) in Fig. 1, as shown in Fig. 4 (c). We extracted from the SEM data the size of the resulting nanogaps as a function of the writing dose, as plotted in Fig. 5. This figure indicates that the metal nanogaps could be lowered to 10nm in size, and should be useful for the detection of single molecules or quantum dots, or single molecule devices.

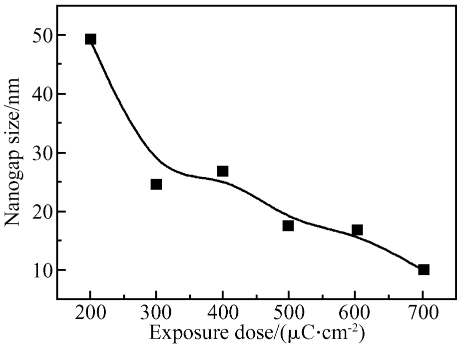

Fig. 5 Plot of the relation between the size of the resulting nanogaps and the writing dose in the range from 200 to 700μC $\cdot$ cm^{-2} The line is a guide for the eye.

4 Summary

We described an alternative fabrication method for the nanogaps down to 10nm and their arrays resulting from the precursor nanoconnections prepared by the extended proximity effect in EBL processes. The size of the nanogaps can be controllable and varied in the range of 10～50nm by adjusting the writing dose. We expect that the as-prepared metal nanogaps will open up applications in investigations of single molecules and single electron nanodevices.

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拓展临近效应由纳米连接制备亚 20nm 金属 Nanogaps*

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摘要： 描述了一种拓展电子束光刻中的临近效应来制备特征尺寸在亚 20nm 的金属 Nanogap 的方法.结合图形转移过程（如去胶等），利用临近效应灵活有效地制备了金属（如 Au 或者 Ag 等）Nanogap 结构及其阵列.采用 GDSII 软件设计图形，以电子束光刻为手段制备 Nanogap 的原始纳米连接图形，然后通过去胶过程获得金属的 Nanogap.另外，通过控制电子束光刻的剂量，能够把 Nanogap 的尺寸降低到约 10nm.

关键词： 金属 nanogap；纳米构筑技术；临近效应；电子束光刻

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