Mechanical frictional scanning probe lithography of TMDCs

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Abstract. In this work, we investigate mechanical scanning probe lithography (SPL) of thick MoSe₂ flakes. The conventional technique faces difficulties in processing the thick samples due to cantilever twisting that leads to the growth of a number of defects and artifacts that decrease spatial resolution. In course of this work, we proposed the approach of frictional-SPL based on small pressure force and many repetitions of lithographic patterns. This approach allows to avoid the formation of remarkable defects and maintain high spatial resolution. By frictional-SPL, we processed thick MoSe₂ flakes (up to 40 nm thick) with the highest resolution down to 20 nm. The results of this work show that frictional-SPL is an effective method of resistless lithography suitable for fabricating nanodevices based on transition metal dichalcogenides (TMDC) materials.

1. Introduction

Scanning Probe Lithography (SPL) is a well-known method of resistless lithography. It can be used for processing both classical materials (such as Si[1], SiC[2], etc.) and low-dimensional materials (including graphene[3], TMDCs[4,5], etc.) that are sensitive to surface adsorbates or any chemical treatment. Nowadays, these materials attract much attention due to their unique properties, and high-resolution resistless methods of lithography are in great demand[6]. SPL provides a broad variety of techniques such as mechanical-SPL, thermal-SPL, oxidation-SPL, etc[7]. Many of them are well-studied and applied for creating research samples and devices. For example, it has been shown that oxidative-SPL can be used for lithography of graphene[3], MoSe₂[5], WSe₂[4], etc. with resolution down to 10 nm. However, this technique may be poorly suitable for processing materials sensitive to oxygen or high humidity such as InSe[8], GaSe[9], etc. While mechanical-SPL implemented under vacuum may be a better option for these materials. Regardless, mechanical-SPL is rarely used per se. Most often it is combined with thermal-/dip pen-SPL, applied for creating patterns on a resist, or used for lithography on the soft organic materials[10]. Although mechanical-SPL has been used for patterning thin graphene layers, it is well known that many artifacts, drags, wrinkles, etc. may appear during the mechanical lithography process[11,12]. These artifacts significantly decrease the spatial resolution of this technique and limit its applicability for processing relatively thick van der Waals heterostructures because the thicker a structure is, the greater force is needed to be applied, and the more artifacts appear.

This work aimed to investigate the use of mechanical scanning probe lithography for patterning layered materials, namely MoSe₂. Here, we propose a novel approach to the implementation of mechanical-SPL based on friction which allows to minimize the emergence of artifacts, achieve spatial resolution down to 20 nm, and pattern flakes with thickness up to 40 nm.
2. Samples and Methods
An experimental sample consisted of a Si substrate covered with 50 nm of gold. MoSe\textsubscript{2} flakes were obtained via micromechanical exfoliation (i.e., scotch-tape method) and transferred on the substrate surface. Experiments were performed on a Ntegra Aura (NT-MDT) scanning probe microscope using DCP (NT-MDT) probes with a curvature radius of 100 nm and a spring constant of 35-85 N/m. The scheme of the experiment is presented in Figure 1.

![Figure 1](image)

**Figure 1** – The scheme of the mechanical scanning probe lithography at various pressures.

Figure 1 shows three typical cases during mechanical-SPL of a layered thick sample. The small pressure of a probe on a surface can provide a smooth but shallow trench. To increase the depth of the pit, pressure clearly needs to be raised. However, the rise of applied force leads to the twisting of the cantilever and the formation of ripped trenches and defects (see medium pressure and intense pressure in figure 1). To pattern thick samples without losing locality, we propose the following approach called frictional-SPL. It consists in multiple repetitions of a lithographic pattern using small pressure on the sample. This approach allows avoiding the cantilever twisting that prevents the formation of artifacts and makes it possible to maintain high resolution even in the case of thick samples. Lithography is carried out through gradual rubbing out material from the surface (not through cutting). Moreover, this approach provides better control over the depth of a pattern.

It is also worth noticing that rigid cantilevers ensure a more stable lithography process due to the difficulty of twisting. For this reason, we use DCP probes with a spring constant of 35-85 N/m for these experiments.

3. Results and Discussions
Figure 2 shows the results of the frictional mechanical-SPL approach investigation. First of all, simple lines were made using various pressure of the probe on the sample surface (see figure 2a). With an increase in the applied force, the number of artifacts and defects of lithography grew. Even using 11 µN led to the loss of locality, providing only the 15 nm depth trench, while using 18 µN resulted in delamination of large sheets of MoSe\textsubscript{2}. Then we identified the threshold of defect manifestation and choose a few values of pressure that lead to smooth lithography (i.e., 5 µN and 7.5 µN). Figure 2b shows the results of the experiment on frictional lithography at various numbers of repetitions and the constant pressure of 5 µN. As can be seen, an increase in the number of repetitions leads to deepening of the pits and does not result in an increase in the number of artifacts and defects. Results of experiments were plotted on the graph (see figure 2c).
Figure 2 – Investigation of mechanical-SPL. (a) – AFM image of the area processed via classical mechanical-SPL using pressure forces from 2 µN to 18 µN. Yellow arrows indicate defects and artifacts; (b) – AFM image of the area processed via frictional-SPL using constant pressure force 5 µN and numbers of iterations from 1 to 100; (c) – The dependence of the depth of pits on the applied pressure force and the number of iterations.

Figure 2c demonstrates data on the depth of pits obtained using classical mechanical-SPL at various applied forces (red curve) and proposed frictional-SPL at various numbers of iterations and constant forces (blue curves). As you can see, the increase in applied force leads to the drastic, non-monotonous, unpredictable increase in the depth of the pit. While the frictional technique provides the smooth, monotonous dependencies of the depth of pits on the number of iterations. Using only 7.5 µN, we could achieve the same depth as using 18 µN, but without any defects or artifacts.

The proposed frictional-SPL technique makes it possible to perform high-resolution lithography. In figure 3 below we demonstrate the use of this approach to create various patterns on MoSe2.

Figure 3 – AFM images of the examples of patterns made by frictional-SPL. (a) – concentric circles with radius from 650 nm to 90 nm; (b) – 20 nm nanoconstriction; (c) – handwriting.

As you can in figure 3, any pattern may be realized using frictional-SPL. The highest spatial resolution we have achieved is 20 nm in the nanoconstriction (see figure 3b). It should be taken into account that DPC probes with a curvature radius of 100 nm were used in these experiments. Using sharper probes certainly will allow achieving higher spatial resolution.
4. Conclusions
In this study, we investigated frictional-SPL of thick MoSe$_2$ flakes. It was shown that the conventional way to deepen the lithography pattern, i.e. the increase of pressure force, leads to the growth of a number of defects and artifacts that decrease spatial resolution and even can destroy the sample. In course of this work, we proposed the approach of frictional-SPL based on small pressure force and many repetitions of lithographic patterns. It was shown that this approach allows to avoid the formation of remarkable defects and maintain high spatial resolution. To demonstrate the possibilities of this method, the flakes with thickness up to 40 nm were processed. We showed the possibility to create any patterns even the handwriting by frictional-SPL. The highest achieved resolution is 20 nm. Thus, the results of this work show that frictional-SPL is an effective method of resistless lithography suitable for fabricating nanodevices based on sensitive layered materials.

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