Mechanical scanning probe lithography of nanophotonic devices based on multilayer TMDCs

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Abstract. In this work, we demonstrate the possibility of using mechanical Scanning probe lithography (m-SPL) for fabricating nanophotonic devices based on multilayered transition metal dichalcogenides (TMDCs). By m-SPM, we created a nanophotonic resonator from a 70-nm thick MoSe₂ flake transferred on Si/Au substrate. The optical properties of the created structure were investigated by measuring microphotoluminescence. The resonator exhibits four resonance PL peaks shifted in the long-wavelength area from the flake PL peak. Thus, here we demonstrate that m-SPL is a high-precision lithography method suitable for creating nanophotonic devices based on multilayered TMDCs.

1. Introduction

After graphene, Transition Metal Dichalcogenides (TMDCs) seem to be the most studied layered materials. This is due to the unique optical and electronic properties of their monolayers such as direct band gap[1], large exciton binding energy[2], valley polarization[3], etc. Moreover, such strong interest in monolayers of these materials encounters researchers to take a fresh look at the properties of bulk TMDCs. Multilayer TMDCs are attractive to nanophotonic applications owing to the high values of the refractive index and its strong anisotropy caused by the layered structure[4]. These properties allow the creation of optical waveguides and light resonators based on multilayer TMDCs. However, these materials are highly sensitive to surface conditions, and some of them can degrade even in ambient conditions[5]. Thus, the traditional methods of lithography involving resist usage and its chemical treatment are poorly suited for creating TMDC nanophotonic structures. Nevertheless, Scanning Probe Microscopy (SPM) can offer several resistless techniques of lithography suitable for creating TMDC nanophotonic devices[6]. For example, oxidative Scanning Probe Lithography (o-SPL) has established itself as an effective resistless method of lithography of layered materials with a resolution down to 10 nm. It has already been successfully used for graphene[7], MoSe₂[8], WSe₂[9], etc. However, we suggest that for materials that are sensitive to ambient conditions and humidity, mechanical-SPL is more suitable. Besides, applying mechanical-SPL for TMDCs is barely studied. This method consists in the mechanical pressure of a probe on a sample with force enabling partial destruction of the surface (puncture, cutting, scratching, etc.). Thus, this lithography technique does not require any resist nor chemical treatment and can be performed in vacuum conditions. Our work aimed to demonstrate the possibility of creating nanophotonic devices (resonators) based on multilayer MoSe₂ by mechanical-SPL.
2. Samples and methods

For the creation of nanophotonic devices, the following structure was prepared. MoSe\(_2\) multilayer flakes obtained by adhesive tape exfoliation were transferred on the Si substrate covered with 50 nm of gold. The experiment was carried out on a Ntegra Aura (NT-MDT) SPM using HA_NC_DCP probes with a tip curvature radius of 100 nm, a spring constant of 35-85 N/m, and a resonant frequency of 350-650 kHz. Ntegra Aura provides a thermal drift of about 10 nm/℃ and a mechanical drift of about a dozen of nm per hour, which ensures the high stability of the sample during the fabrication of a resonator. The scheme of Mechanical Scanning Probe Lithography (m-SPL) is shown in Figure 1.

![Diagram of m-SPL](image)

**Figure 1.** The scheme of Mechanical Scanning Probe Lithography.

After selecting a flake of suitable sizes (with a thickness comparable to the wavelength of visible light in MoSe\(_2\)), m-SPL was used to create nanophotonic structures. During m-SPL the probe passes the chosen lithographic pattern with given pressure on the surface. Mechanical interaction of the rigid diamond-coated probe with the relatively fragile surface of TMDC flake results in the erasing of surface TMDC layers. A few repetitions of such procedure can provide complete deleting of TMDC flake in chosen areas. In our experiments, the hard probes with a spring constant of 35-85 N/m were used to prevent axial twisting of the cantilever, which takes place using soft probes and leads to a decrease in lithography resolution.

Optical properties were studied by measuring microphotoluminescence (PL) spectra. The experiments were carried out at room temperature (300 K) using Horiba Jobin Yvon T64000. The measurements were performed with continuous-wave (cw) excitation using the 532 nm laser line of a Nd: YAG laser. We used a Mitutoyo 100 × NIR (NA = 0.90) long working-distance objective lens to focus the incident beam into a spot of ~ 0.7 μm diameter.

Multilayer TMDC flakes can exhibit photoluminescence at high excitation densities. An excitation power of 4 mW was used here.

3. Results and discussions

Figure 2(a-c) shows the process of investigated nanophotonic device creation. To create a resonator, a 70 nm thick flake was chosen (Figure 2a). First of all, a dozen of lithography iterations were performed to reveal a degree of interaction between the probe and the flake and to determine the suitable pressure force (Figure 2b). Then, a few hundreds of iterations of lithography provided the creation of the structure with the given size and shape. Figure 2c shows the finished resonator with a thickness of 70±2nm and a diameter of 1500±30 nm. The gap between the created structure and the flake is of 900 nm, which protects the modes in the resonator from leaking in the flake. Figure 2d demonstrates the height profile of the structure. As you can see, the level of lithography is lower than the level of the Au sublayer. This indicates that the resonator is fully separated from the flake, and
there are no thin MoSe$_2$ layers on the floor of the trench. The optical properties of the created structure were investigated by Photoluminescence spectroscopy (PL).

![Figure 2](image)

**Figure 2.** (a-c) Steps of the resonator creation: (a) the chosen flake; (b) determination of lithography parameters; (c) the finished structure. Colored dots show PL measurement areas. (d) The height profile of the structure; (e) Results of PL investigation of the structure.

Figure 2d shows the results of the PL investigation of the structure. The green curve (corresponding to the green dot in Figure 2c) was measured in the pit around the resonator. It can be seen that, apart from the background photoluminescence associated with the plasmon in gold[10], there are no peaks in the investigated wavelength range. The black curve (corresponding to the black dot in Figure 2c) was measured on the flake close to the resonator. This spectrum demonstrates a peak related to the photoluminescence of the multilayered MoSe$_2$ flake. The red curve (corresponding to the red dot in Figure 2c) was measured on the resonator. As can be seen, the spectrum of the resonator significantly differs from the one of the flake. Four peaks shifted in the long-wavelength area unambiguously indicate the presence of resonance in the created structure.

### 4. Conclusions

To conclude, in this work, the possibility of using mechanical scanning probe lithography for processing multilayered TMDCs was demonstrated. In the course of our work, we created a nanophotonic resonator with a thickness of 70 nm and a diameter of 1500 nm by m-SPL. The resonator exhibits four PL peaks shifted in the long-wavelength area from the flake PL peak that unambiguously indicates the presence of resonance in the created structure. Thus, it was demonstrated that m-SPL can be considered an effective method of creating nanophotonic devices based on multilayered TMDCs.

### References

[1] Mak K F, Lee C, Hone J, Shan J and Heinz T F 2010 Atomically thin MoS$_2$: a new direct-gap semiconductor *Phys. Rev. Lett.* **105** 136805
[2] Wang G, Gerber I, Bouet L, Lagarde D, Balocchi A, Vidal M, Amand T, Marie X and Urbaszek B 2015 Exciton states in monolayer MoSe2: impact on interband transitions 2D Mater. 2 045005

[3] Gong Z, Liu G-B, Yu H, Xiao D, Cui X, Xu X and Yao W 2013 Magnetoelectric effects and valley-controlled spin quantum gates in transition metal dichalcogenide bilayers Nat. Commun. 4 1–6

[4] Ermolaev G, Grudinin D, Stebunov Y, Voronin K, Kravets V, Duan J, Mazitov A, Tselikov G, Bylinkin A, Yakubovsky D, and others 2021 Giant optical anisotropy in transition metal dichalcogenides for next-generation photonics Nat. Commun. 12 1–8

[5] Diaz H C, Chaghi R, Ma Y and Batzill M 2015 Molecular beam epitaxy of the van der Waals heterostructure MoTe2 on MoS2: phase, thermal, and chemical stability 2D Mater. 2 044010

[6] Ryu Y K and Garcia R 2017 Advanced oxidation scanning probe lithography Nanotechnology 28 142003

[7] Alekseev P, Borodin B, Dunaevskii M, Smirnov A, Davydov V Y, Lebedev S and Lebedev A 2018 Local Anodic Oxidation of Graphene Layers on SiC Tech. Phys. Lett. 44 381–38

[8] Borodin B R, Benimetskiy F and Alekseev P A 2021 Study of local anodic oxidation regimes in MoSe2 Nanotechnology 32 155304

[9] Dago A I, Ryu Y K and Garcia R 2016 Sub-20 nm patterning of thin layer WSe2 by scanning probe lithography Appl. Phys. Lett. 109 163103

[10] Sivun D, Vidal C, Munkhbat B, Arnold N, Klar T A and Hrelescu C 2016 Anticorrelation of photoluminiscence from gold nanoparticle dimers with hot-spot intensity Nano Lett. 16 7203–9