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RESEARCH ARTICLE

Fabrication of two-dimensional semiconductors on the surface of ferroelectric films by means of gold-assisted mechanical exfoliation

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Abstract

Objectives. The aim of this study is to develop and demonstrate an effective method for obtaining large-area, high-quality monolayers of molybdenum disulfide (MoS_2) on the surface of ferroelectric lead zirconate titanate (PZT) films which exhibit pronounced granularity and texturing. Conventional mechanical exfoliation techniques are inefficient for transferring two-dimensional materials onto nonplanar surfaces. This is due to local height variations and substrate granularity which hinder the formation of continuous monolayers and high-defect-density transferred structures. A particular challenge is the transfer onto functional substrates with surface topography characterized by heterogeneities ranging from tens of nanometers to micrometers.

Methods. A gold-assisted exfoliation (GAE) method was employed, including: magnetron sputtering of a 50 nm gold film; mechanical delamination of monolayers using thermally cleavable tape; and subsequent gold etching. The characterization was performed using X-ray diffraction, optical confocal microscopy, atomic force microscopy, and second harmonic generation techniques. The efficiency of the transfer process was compared for Si/SiO_2 and PZT substrates.

Results. MoS_2 crystallites with areas up to 3000 μm^2 were obtained on PZT and over 65000 μm^2 on standard Si/SiO_2 substrates, both of which exhibit minimal defect densities. Conventional mechanical exfoliation is shown to be unable to ensure transfer onto textured surfaces, whereas the GAE method preserves the monolayer character of the transferred crystallites even on nonplanar substrates.

Conclusions. This work demonstrates for the first time the possibility of obtaining large-area, high-quality MoS_2 monolayers on substrates with pronounced grainy and textured structures, such as ferroelectric PZT films, using the gold-assisted exfoliation method. The work also shows that gold-assisted exfoliation is an effective technique for fabricating extended two-dimensional films with controlled morphological and structural properties, including on substrates previously considered unsuitable for such applications.

Keywords: two-dimensional materials, molybdenum disulfide, gold-assisted exfoliation, ferroelectric thin films, lead zirconate titanate, mechanical exfoliation, nanostructures, FeFET

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НАУЧНАЯ СТАТЬЯ

Создание двумерных полупроводников на поверхности сегнетоэлектрических пленок методом механической эксфолиации при помощи золота

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Резюме

Цели. Цель работы заключается в разработке и демонстрации эффективного метода получения протяженных и высококачественных монослоев дисульфида молибдена (MoS_2) на поверхности сегнетоэлектрических пленок цирконата-титаната свинца (ЦТС) с выраженной зернистой и текстурированной структурой. Стандартные методы механической эксфолиации оказываются неэффективными для переноса двумерных материалов на неровные поверхности из-за локальных перепадов высоты и зернистости подложки, что приводит к невозможности формирования протяженных монослоев и высокой плотности дефектов в переносимых структурах. Особую сложность представляет перенос на функциональные подложки с рельефом поверхности, характеризующимся неоднородностью на масштабах от десятков нанометров до микрометров.

Методы. Использован метод золото-ассистированной эксфолиации (gold-assisted exfoliation, GAE), включающий магнетронное напыление золотой пленки толщиной 50 нм, механическое отделение монослоев с помощью терморасщепляемого скотча и последующее травление золота. Характеризация проведена методами рентгеновской дифракции, оптической конфокальной микроскопии, атомно-силовой микроскопии и генерации второй оптической гармоники. Сравнение эффективности переноса выполнено на подложках кремния/оксида кремния (Si/SiO_2) и ЦТС.

Результаты. Получены кристаллиты MoS_2 площадью до 3000 мкм^2 на ЦТС и свыше 65000 мкм^2 – на стандартных подложках Si/SiO_2 при минимальной плотности дефектов. Показано, что стандартная механическая эксфолиация не обеспечивает перенос на текстурированные поверхности, тогда как GAE сохраняет монослойность переносимых кристаллитов даже на неровных подложках.

Выводы. Впервые продемонстрирована возможность получения протяженных и высококачественных монослоев MoS_2 на подложках с выраженной зернистой и текстурированной структурой, таких как сегнетоэлектрические пленки ЦТС, с помощью метода золото-ассистированной эксфолиации. Показано, что золото-ассистированная эксфолиация представляет собой эффективный метод для создания протяженных двумерных пленок с контролируемыми морфологическими и структурными характеристиками, в т.ч. на подложках, ранее считавшихся непригодными для подобных задач.

Ключевые слова: двумерные материалы, дисульфид молибдена, золото-ассистированная эксфолиация, сегнетоэлектрические пленки, цирконат-титанат свинца, механическая эксфолиация, наноструктуры, FeFET

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INTRODUCTION

Over the past two decades, two-dimensional (2D) materials such as graphene, molybdenum disulfide, and hexagonal boron nitride have become the subject of intensive research due to their unique physical, optical, and electronic properties, fundamentally different from those of their bulk analogs [1, 2]. These properties are due to the quantum size effect, high anisotropy, increased specific surface area, and changes in the electronic structure upon going to atomically thin layers. 2D materials in particular exhibit unusual quantum phenomena such as the quantum Hall effect, strong fluorescence in MoS_2 monolayers, and ultra-high charge carrier mobility. This opens new horizons for fundamental research in condensed matter physics and materials science [3].

Thanks to these properties, two-dimensional materials are being considered as platforms for creating new generations of electronic, optoelectronic, energy, and sensor devices. However, the practical realization of their promise is closely linked to the ability to produce high-quality, defect-free, and extended-area monolayers, as well as their integration into modern micro- and nanoelectronic technologies. Until recently, the synthesis of such layers and their transfer to functional substrates was a serious technological challenge, significantly hindering both in-depth research into fundamental principles and the development of applied solutions.

There are several methods for producing two-dimensional materials, each with its own advantages and limitations. The most common approaches include mechanical exfoliation, liquid exfoliation, and chemical vapor deposition (CVD). Mechanical exfoliation, first used to synthesize graphene, remains the benchmark method for producing high-quality samples with low defect density [4]. However, its scalability is limited by small lateral dimensions. Liquid exfoliation enables the mass production of materials but leads to crystal fragmentation and contamination. Chemical vapor deposition enables the synthesis of large-area films, but the defect density in such materials often exceeds 10^{12}–10^{13} cm^{-2} [5].

One possible solution to the problem of creating layers with the desired characteristics is the gold-assisted exfoliation (GAE) method which has been relatively recently developed. This method combines the advantages of mechanical exfoliation and scalability, enabling the production of single crystals up to a centimeter in size [6–8], while the defect density in the resulting layers is minimal and can reach $\sim 3 \cdot 10^{11}$ cm^{-2} [9]. Thus, GAE is the preferred method for applications where high structural perfection and surface cleanliness are critical.

Despite significant progress in producing 2D materials, integrating high-quality 2D crystals into real devices still poses significant technological challenges. One of the key challenges in modern electronics is the creation of functional structures in which 2D layers are transferred onto substrates with distinct surface topography. These include such films as ferroelectric lead zirconate titanate ($Pb(Zr_{0.48}Ti_{0.52})O_3$, PZT) films used in ferroelectric field-effect transistor (FeFET) architectures and other memristor devices. However, standard mechanical exfoliation methods are ineffective for transferring 2D materials to such uneven or textured surfaces. Localized differences in height and substrate graininess dramatically reduce the contact area between the 2D crystal and the substrate, making it impossible to form extended and continuous monolayers. As a result, attempts to use classical approaches lead either to a complete absence of transfer or to the formation of highly fragmented and defective regions unsuitable for practical application.

In order to overcome these limitations, new methods must be developed to ensure selective and delicate transfer of 2D materials, even onto complex substrates with pronounced microroughness. One of the most promising solutions is the GAE method mentioned above. However, its application to textured ferroelectric films has remained largely unexplored to date. Given that such structures are essential for the implementation of modern FeFET and memristor devices, demonstrating the feasibility of efficient transfer of 2D materials onto PZT and similar surfaces is of fundamental and applied importance.

The main objective of this study is to experimentally demonstrate that gold-assisted exfoliation enables the formation of extended, high-quality monolayers of 2D materials not only on standard flat substrates, but also on functional ferroelectric films with distinct topography. This study compares the transfer efficiency of SiO_2 and PZT, and analyzes the morphology, structural perfection, and area of the resulting crystallites. Particular attention is paid to the comparison with the results obtained using classical mechanical exfoliation. As this study shows, this does not, however, ensure the transfer of 2D materials onto uneven substrates. Thus, the results of this study not only expand our fundamental understanding of the mechanisms of interaction between 2D crystals and textured surfaces, but also open up new possibilities for integrating 2D materials into promising electronic and spintronic devices.

1. MATERIALS AND METHODS

1.1. Methodology for creating two-dimensional films

Molybdenum disulfide (MoS_2) was chosen as the two-dimensional semiconductor material for creating monolayers. This is due to two key factors: its widespread use in modern research; and its strong chemical interaction with gold [10]. Gold atoms, which possess vacant *d*-orbitals, effectively interact with the lone electron pairs of sulfur atoms, forming strong covalent bonds. This interaction ensures the high selectivity of the GAE method with respect to sulfur-containing compounds, making MoS_2 the optimal material for developing this technique.

In order to verify the developed method, experiments were conducted in two stages. In the first stage, a standard silicon/silicon oxide (Si/SiO_2) structure, traditionally used in mechanical exfoliation [11, 12], was used as a substrate for the formation of reference quasi-two-dimensional crystallites. In the second stage, the gold-assisted exfoliation technology was adapted for deposition onto a textured ferroelectric PZT film. This allowed for the applicability of the method for creating two-dimensional structures on functional materials with a non-uniform surface to be evaluated.

PZT films ($Pb(Zr_{0.48}Ti_{0.52})O_3$) were formed from a film-forming solution prepared by the sol–gel method using a complex of zirconium isopropoxide monosolvate ($Zr[OCH(CH_3)_2]_4(CH_3)_2CHOH$, Sigma-Aldrich, USA), titanium tetraisopropoxide ($Ti[O(CH_3)_2CH]_4$, Sigma-Aldrich) and lead acetate ($Pb(CH_3COO)_2$) synthesized from lead oxide (PbO, Sigma-Aldrich) according to a previously developed technique [13]. The PZT solution was applied layer by layer (6 layers, ~220 nm) by spin coating on platinized silicon substrates with the following structure: Pt (~150 nm)–TiO_2 (~10 nm)–SiO_2 (~300 nm)–Si (~800 μm). After spin coating (2500 rpm), each layer was dried for 5 min with an infrared lamp in pulsed mode at a temperature of ~200°C and annealed for 15 min at a temperature of 400°C in an SNOL 1.6.2.5.1/10-IZM muffle furnace (Termiks, Russia). After heat treatment of the sixth layer, the sample was crystallized for 10 min using an AS-One 150 fast annealing unit (Annealsys, France) at a temperature of 650°C (heating rate ~15°C/s).

The process of creating single-crystal samples involves five key steps which are detailed in Fig. 1. In the first step, a bulk molybdenum disulfide crystal is separated layer by layer using adhesive tape (e.g., silicone tape). This step enables the production of multilayer fragments with thicknesses ranging from a few nanometers to microns. In the second step, a 50-nm-thick gold film is deposited onto the pre-treated multilayer MoS_2 crystallites using a VUP-5 magnetron setup (Russia). The process is carried out in a vacuum chamber at a power of 100 W and an argon pressure of $5 \cdot 10^{-3}$ Torr. In the third step, thermal release tape (TRT) is tightly pressed onto the gold surface. After fixation, the tape is carefully peeled off from the original substrate, thus capturing the top MoS_2 layer along with the gold film. The Au–MoS_2 adhesive force exceeds the interlayer van der Waals bonds in MoS_2, enabling selective detachment of the monolayer. In the fourth step, the resulting TRT–Au–MoS_2 structure is transferred to a Si/SiO_2 substrate, in order for the MoS_2 layer to remain in contact with the substrate. The substrate is then heated on a hot plate at 130–150°C for 1–2 min. This weakens the adhesion of the TRT tape, enabling its removal without leaving a trace. In the final step, the gold film is removed by immersion in an etching solution (4 g KI + + 1 g I_2 in 40 mL H_2O) for 3–5 min at room temperature. MoS_2 demonstrates the chemical inertness to this reagent due to its covalent S–Mo–S structure. After the etching is complete, the surface is cleaned for 10 min with acetone to remove any remaining etchant, followed by a rinse with isopropyl alcohol. This procedure ensures the complete removal of contaminants and reagent residues, facilitating the formation of clean, high-quality 2D semiconductor monolayers with large lateral dimensions.

1.2. Research methods

X-ray diffraction was used to analyze the crystal structure, allowing us to determine interplanar spacings and structural symmetry. This is necessary, in order to verify the phase purity of bulk molybdenum disulfide and assess the quality of the PZT films, which served as the basis for creating the two-dimensional films. This approach allows us to confirm the material compliance with the specified characteristics and their suitability for further research.

Diffraction patterns were obtained using a DRON-8T diffractometer (Burevestnik, Russia) at a voltage of 40 kV and a current of 20 mA. Due to the limitations of the method, only bulk MoS_2 samples and polycrystalline

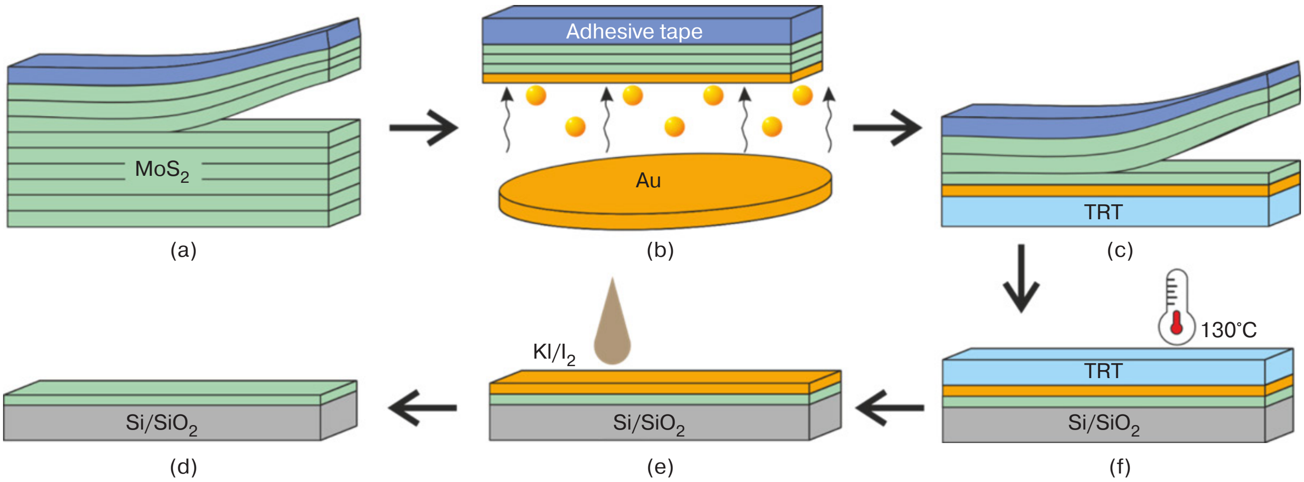


Fig. 1. Process of creating monolayer MoS$_2$ films:
(a) mechanical exfoliation of MoS$_2$; (b) magnetron sputtering of gold on MoS$_2$;
(c) application of TRT tape to gold and subsequent exfoliation of MoS$_2$; (d) removal of TRT tape by heating;
(e) etching of gold with a KI/I$_2$ solution; (f) the finished monolayer on a Si/SiO$_2$ substrate

PZT films were studied, since the small thickness of two-dimensional films significantly complicates the separation of the diffraction signal from the substrate.

Following GAE exfoliation, standard optical confocal microscopy was used to detect monolayer MoS$_2$ crystallites on the substrate surface. This approach is optimal for rapid preliminary analysis, enabling the determination of crystallite thickness, size, and the presence of defects [8]. An Alpha 300s+ optical microscope (WITec, Germany) with high-aperture objectives and color filters was used. This provides enhanced image contrast for monolayer crystallites, critical when working with atomically thin materials. This method effectively identifies promising areas for subsequent detailed study using other analytical methods.

Atomic force microscopy (AFM), which complements optical data, was used to accurately assess the topography of two-dimensional materials. Atomic force microscopy provides a three-dimensional image of the surface with a resolution of up to nanometers, enabling the detection of defects such as cracks, dislocations, and roughness. This is especially important for quality control of the resulting two-dimensional films and the assessment of the influence of the substrate on their morphology [14]. All measurements were performed using an Ntegra Prima scanning probe microscopy system (NT-MDT, Russia) in contact mode. Rigid probes of the Etalon series with a spring constant of 12 N/m were used, ensuring an optimal balance between resolution and the integrity of the studied samples.

The standard fluorescence microscopy-spectroscopy technique was not used in this study. This is due to the complete signal quenching caused by residual gold nanostructures after the GAE technique. Even very small amounts of gold (Au), retained on the substrate surface or near two-dimensional crystallites, act as effective nonradiative recombination centers. This is due to two main mechanisms of luminescence quenching. The first mechanism is the charge quenching effect: electron transfer from MoS$_2$ to Au leads to *p*-doping of the material and suppression of exciton luminescence. The second mechanism is resonant energy transfer: at distances less than 10 nm between Au and MoS$_2$ non-radiative energy transfer from excitons to plasmonic modes of gold occurs [15, 16]. In addition, this technique is not applicable to samples thicker than one monolayer [17].

For this reason, second harmonic generation (SHG) was additionally used to study structural symmetry, crystallographic orientation, and defects in two-dimensional materials. This nonlinear optical method is based on the conversion of two photons of exciting radiation into one photon with doubled frequency in non-centrosymmetric media. SHG is particularly effective for studying thin MoS$_2$ layers, since the signal intensity depends significantly on the number of layers and their polytype. In regions with broken crystal symmetry, including edges, defects, and transitions between domains, a significant enhancement of the non-linear response is observed, ensuring high sensitivity of the method to the structural features of two-dimensional crystallites.

The experimental setup used a TiF-DP 60 femtosecond titanium:sapphire laser (Avesta, Russia) with a pulse duration of 20–60 fs and a repetition rate of 80 MHz. The femtosecond pulses provided high peak power with low average power, enabling an intense nonlinear response without thermal damage to the sample. A modified WITec Alpha 300S+ confocal microscope was also used for precise sample positioning and scanning.

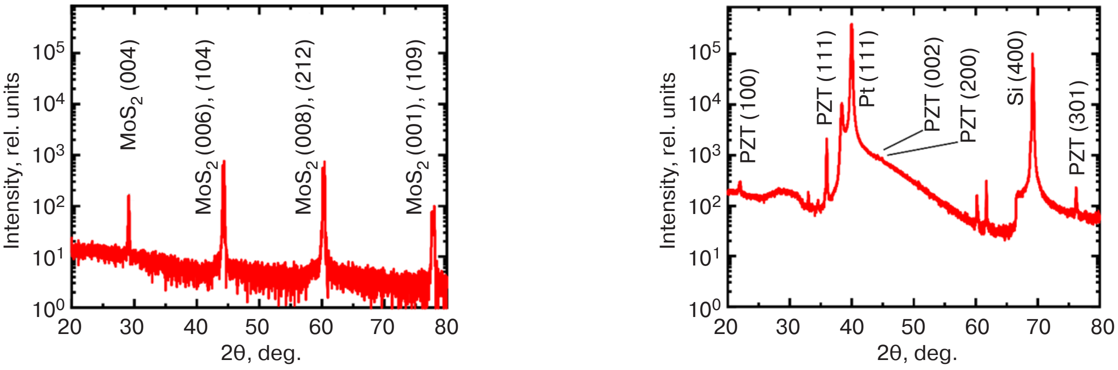
2. RESULTS

In order to characterize the crystal structure and phase composition of the starting materials, X-ray diffraction studies were carried out in the 2θ-scanning mode. The results are presented in Fig. 2. The diffraction pattern of the bulk MoS_2 crystal demonstrates clearly defined reflections (004), (006), and (008), indicating a high degree of crystallinity and a preferential orientation of the crystallites along the axis perpendicular to the plane of the layers. The nature of the diffraction pattern indicates a hexagonal structure of the 2H or 3R polytype of MoS_2, fully consistent with the literature data [18, 19]. Analysis of the diffraction pattern of the PZT film revealed the presence of characteristic peaks (100), (111), (002), (200), and (301), confirming the formation of the perovskite phase with a preferential orientation in the (111) plane [20]. The formation of the perovskite-like AVO_3 structure in PZT provides the emergence of ferroelectric properties. This is due to the displacement of Ti^{4+}/Zr^{4+} and Pb^{2+} cations from centrosymmetric positions in the oxygen octahedra, which disrupts the inversion symmetry of the crystal and creates spontaneous polarization [21]. The possibility of reorienting this polarization by an external electric field determines the ferroelectric behavior of the material, as confirmed by characteristic hysteresis loops in the polarization-electric field dependences [22]. The low-intensity reflections corresponding to the Pt and Si phases are associated with the multilayer structure of the substrate used. They do not affect the functional properties of the ferroelectric film. Quantitative analysis of the half-width of the diffraction lines indicates the absence of significant microstrains and structural defects in the studied materials, confirming their high crystalline perfection and structural homogeneity. These results indicate the suitability of the selected materials for further use as feedstock and functional substrate in the creation of heterostructures by gold-assisted exfoliation.

In order to analyze the influence of the surface parameters of the used substrates on the efficiency of the GAE technique, the surfaces of both substrates—Si/SiO_2 and PZT—were analyzed using the AFM technique. The resulting three-dimensional maps of the substrate surfaces are presented in Fig. 3. As can be clearly seen from the results obtained, PZT has significantly greater surface inhomogeneities due to the characteristics of grain growth during crystallization. PZT films obtained by the sol–gel method are characterized by the formation of a polycrystalline structure with pronounced grain boundaries, leading to the formation of a textured surface. Further analysis of the results obtained revealed that PZT has a roughness of about $1.090 \cdot 10^{-9}$ m, while for the Si/SiO_2 substrate this parameter is $5.073 \cdot 10^{-10}$ m.

Thus, the height differences on the PZT substrate surface are comparable to or even exceed the thickness of the 2D MoS_2 (films deposited on it (~0.7 nm for a monolayer), leading to a significant reduction in the contact area between the substrate and the film. Non-uniform contact between the materials negatively impacts the van der Waals forces responsible for MoS_2 adhesion to the surface. This in turn degrades the deposition quality of 2D crystallites and leads to the formation of defects at the interfaces between the 2D material and the substrate. All this reduces the efficiency of charge transfer and degrades the electronic performance of the devices.

After sample creation, several hundred two-dimensional MoS_2 crystallites were identified on the surfaces of the Si/SiO_2 and PZT substrates using



(a) (b)

Fig. 2. Diffraction patterns of bulk MoS_2 (crystals (a) and PZT substrate (b)

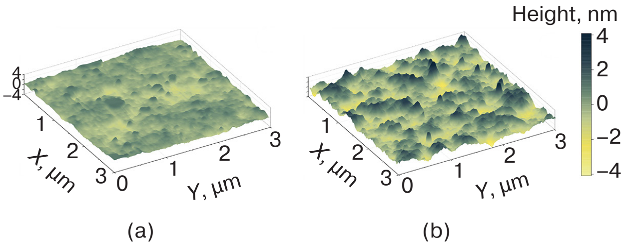


Fig. 3. Three-dimensional surface topography maps of the substrates obtained by AFM: (a) Si/SiO_2 and (b) polycrystalline PZT film

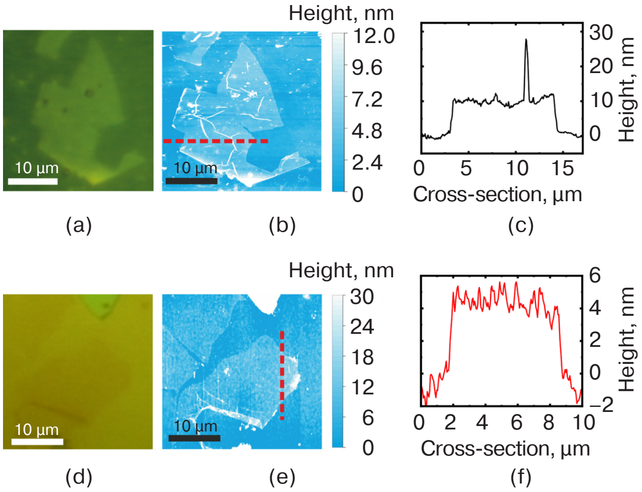


Fig. 4. Results of gold exfoliation: (a) Optical image of a region with MoS_2 crystallites on a Si/SiO_2 substrate; (b) AFM topography of a MoS_2 crystallite on a Si/SiO_2 substrate; (c) The height profile obtained from the section (red dashed line) shown in (b); (d) Optical image of a region with MoS_2 crystallites on a PZT substrate; (e) AFM topography of a MoS_2 crystallite on a PZT substrate; (f) The height profile obtained from the section (red dashed line) shown in (e)

confocal optical microscopy. Representative samples with typical morphological characteristics were selected for detailed analysis: an area of ~200 µm^2, the absence of visible defects, and a uniform distribution over the surface. Figures 4a and 4d show the crystallites for each substrate, selected as standards for the comparative study.

In order to determine the thickness and lateral dimensions of the crystallites, topographic maps were obtained using AFM (Figs. 4b and 4e). The thickness of the film on the Si/SiO_2 substrate was 5 nm, corresponding to ~8 monolayers of MoS_2 with an interlayer distance of 0.65 nm, while on PZT, a thickness of 8 nm (~12 layers) was recorded. The crystallite area was approximately 200 µm^2 for both crystallites.

The average surface roughness of the MoS_2 films was 2.53 nm for both substrates, significantly exceeding the values for the original substrates (0.51 nm for Si/SiO_2 and 1.09 nm for PZT). This increase in roughness is due to the appearance of pronounced morphological irregularities, primarily folds, which are clearly visible in the topographic maps and highlighted in red in Figs. 4c and 4f. The formation of folds is associated with the characteristics of mechanical transfer and the specific application of the GAE technique. In addition, foreign defects caused by residual materials after gold etching can be observed on the crystallite surface. These defects manifest themselves as local inclusions and can be detected both on the substrate and over the entire surface of the monolayer. It is important to note that the grain structure of the PZT substrate is partially visible through the MoS_2 surface, indicating non-uniform contact between the film and the substrate and the presence of regions where MoS_2 appears to be “suspended” above the surface.

Figure 5a shows a scanning electron microscopy (SEM) image of a MoS_2 crystallite on a Si/SiO_2 substrate where characteristic film folds (marked with arrows) formed during mechanical transfer are clearly visible. Topography analysis revealed that the fold height reaches 30 nm with an average length of 5 µm, consistent with the AFM data (Figs. 4b and 4e). Similar SEM measurements were not carried out for the PZT substrate due to the occurrence of local polarization reversal of the ferroelectric film under the

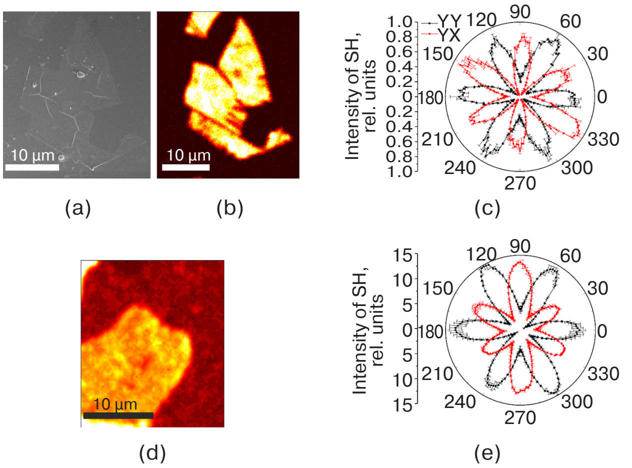
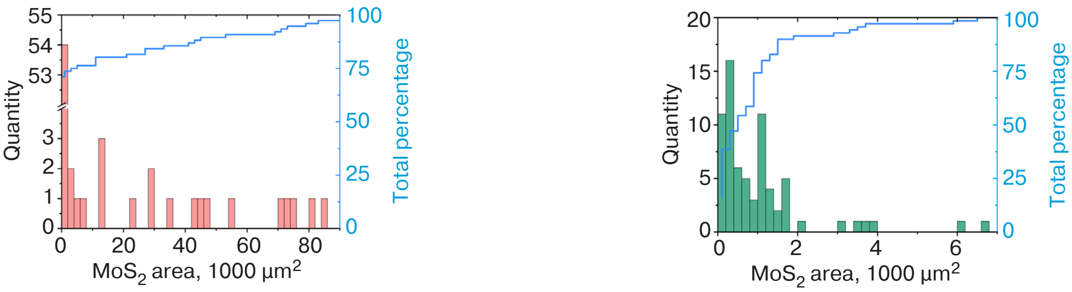


Fig. 5. SEM image of a MoS_2 crystallite on a Si/SiO_2 substrate (a); two-dimensional map of the second harmonic (SH) distribution in quasi-two-dimensional MoS_2 crystallites on Si/SiO_2 and PZT substrates (b), (d); azimuthal dependencies of the SH intensity in MoS_2 crystallites on Si/SiO_2 and PZT substrates (c), (e) in parallel (YY) and crossed (YX) combinations of pump radiation polarization and the selected SH field component



(a) (b)

Fig. 6. Histogram of the area of two-dimensional MoS_2 crystallites on a substrate (a) Si/SiO_2 and (b) PZT

action of the electron beam which significantly distorts the distribution of the second optical harmonic [23]. It has been established experimentally that even short-term (~30 s) exposure to electrons with an energy of 5 kV leads to an irreversible change in the domain structure of PZT, which makes it impossible to correctly compare optical and electron microscopic data.

Spatial maps of the SHG intensity distribution obtained by scanning nonlinear microscopy (Figs. 5b and 5d) enable the reliable identification of morphological defects, including folds and cracks. A local increase in signal intensity is recorded in zones corresponding to such defects. This increase may be due to a violation of local centrosymmetry or to edge effects caused by an increase in nonlinear optical susceptibility near domain boundaries or at the edges of the crystalline region [24], as well as an increase in film thickness.

The six-beam azimuthal dependence of the second optical harmonic intensity in thin MoS_2 (Figs. 5c and 5e) is due to the crystallographic symmetry of the material. In the case of a non-centrosymmetric 3R polytype, as well as with an even number of layers, the structure is characterized by the point symmetry group C_{3v}. This leads to a pronounced six-beam dependence of the second harmonic intensity on the azimuthal rotation angle of the sample [25, 26]. In the experiment, the phases of the intensity maxima coincide at all flake points, indicating the single-crystal nature of the film, and the uniform orientation of the crystallographic axes.

A uniform SHG intensity distribution is observed in most of the MoS_2 film regions studied. This indicates a low density of morphological defects and high crystalline quality of the material. The uniformity of the signal confirms the absence of significant grain boundaries, folds, and cracks over the bulk of the flake, and also indicates a minimal influence of polytypism, in particular, the dominance of the non-centrosymmetric 3R polytype, which makes the largest contribution to the SHG signal.

In order to conduct an objective comparison of the efficiency of forming quasi-two-dimensional films on various substrates, a statistical analysis of the morphological parameters of over 300 created crystallites was performed. One of the key criteria in this case is the area distribution of individual crystallites, since this parameter determines the suitability of the material for further applications and directly depends on both the chosen exfoliation method and the substrate properties.

The area distribution histograms of quasi-two-dimensional MoS_2 crystallites obtained by the GAE method are shown in Fig. 6, demonstrating a significant difference between the Si/SiO_2 (Fig. 6a) and PZT (Fig. 6b) substrates. It is important to note that the statistical sample includes crystallites with an area greater than 0.2 μm^2 and a thickness less than or equal to 7 nm (~10 monolayers), corresponding to the transition region between quantum-well and bulk properties. The lower area threshold was chosen based on technological requirements. Smaller crystallites are unsuitable for the creation of functional devices due to limitations of lithography and the formation of electrode structures.

For Si/SiO_2 substrates, more than 25% of the visualized crystallites have an area exceeding 65000 μm^2, or four orders of magnitude higher than the typical results of conventional exfoliation (<0.5 μm^2) [27]. On the PZT substrate, the distribution is shifted towards the lower values. The area of the largest 25% of crystallites is only 3000 μm^2, associated with increased surface roughness (R_a = 1.09 nm) and, as a result, heterogeneous contact between the film and the substrate. It is fundamentally important that, when using standard mechanical exfoliation, not a single crystallite with an area greater than 1 μm^2 could be detected on the PZT.

Thus, the gold-assisted exfoliation method ensures the formation of extended and high-quality MoS_2 monolayers not only on standard but also on textured substrates, where other approaches prove ineffective.

CONCLUSIONS

For the first time this work demonstrates the possibility of producing extended, high-quality MoS_2 monolayers on substrates with a pronounced granular and textured structure, such as ferroelectric PZT films. The gold-assisted exfoliation method enables the formation of crystallites with an area of up to 3000 μm^2 on PZT and over 65000 μm^2 on standard Si/SiO_2 substrates. This is several orders of magnitude greater than the results achievable using traditional mechanical exfoliation where no crystallites larger than 1 μm^2 can be formed on textured substrates. The crystallinity and homogeneity of the resulting layers were confirmed by AFM and nonlinear optical microscopy.

The results obtained open up new prospects for integrating 2D semiconductors into functional devices such as FeFETs and memristors which require layer transfer onto complex and uneven substrates. The GAE technique can be extrapolated to a wide range of 2D materials and various types of functional surfaces. Thus, gold-assisted exfoliation represents an effective and reproducible tool for creating extended 2D films with controlled morphological and structural characteristics, including on substrates previously considered as unsuitable for such applications.

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Authors' contributions

E.I. Zhemerov—methodology development, literature review, conducting experiments, interpretation and generalization of results, preparation of the initial manuscript draft.

A.A. Guskov—methodology development, conducting experiments, interpretation and generalization of results, preparation of the initial manuscript draft and the article for publication.

E.A. Bulavintseva, **D.S. Seregin**—methodology development, sample preparation.

S.D. Lavrov—creation of research concept and methodology, preparation of the initial manuscript draft, manuscript text editing, general supervision.

All authors have read and approved the published version of the manuscript.

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