Title:	Low power-focused laser irradiation induced controlled nanostructuring of MoS 2 flakes for potential optical and electronic applications
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Abstract: Over the past decades, micro/nanostructures have engrossed a tremendous interest owing to its exceptional features and functional capabilities for extensive development possibilities in the field of nanotechnology. In recent years, the emergence of 2D layered materials has attracted immense research and industrial interest due to their promising layered dependent optical and electronic properties, which opens an unprecedented prospect of next-generation miniaturized and atomic-scale thin nanoelectronic, spintronics, memory devices, bio and chemical sensors platform, etc. Nanostructures of 2D layered materials exhibit an immense scientific value perspective of extending and improving the next generation miniaturized multifunctional devices based on 2D-layered materials. Molybdenum Disulfide (MoS 2) is emerging as a most promising candidate in all 2D layered materials and can be a potential candidate for exploring its nanostructures owing its exotic physical properties such as layered dependent band transition from indirect to direct, valley hall effect, excitonic properties, strong spin-orbit coupling, valley-selective circular dichroism, and stacking sequence-dependent properties. Nanostructuring on MoS 2, i.e., nanoribbons, nanomesh, etc., may open a new prospect of applications in the field of optoelectronic devices, sensing and catalysis due to their unexplored promising optical and electronic properties over the regular 2D configuration. The creation of these complicated nanostructures can be achieved by using standard nanopatterning techniques such as lithography, printing, etc. However, these conventional techniques involve affluent multistep processes to optimize scalability, form factors, and accuracy in the feature size, which hampers the controlled and rapid prototyping of preferred nanostructures and poses challenges in this perspective. In this thesis, we have addressed this key issue, notably the controlled micro/nanostructuring of MoS 2, using a reliable and simple approach to investigate the induced functionalities and exploring its potential applications. However, the identification of layer numbers with accurate thickness is a key prerequisite before carrying out the micro/nanostructuring of MoS 2 . Firstly, we have presented a comprehensive and precise technique for thickness (layer number) determination of 2D flakes by spectroscopic mapping of white light reflection from the flake instead of following the conventional crude way of optical image analysis through RGB filters.This process provides information about the spectral dependency of the optical contrast in the full visible range. It defines the spectral range of filters to be selected for optimized contrast imaging and provides an optical mean for accurate thickness measurements. Further, we have discussed the crucial role of selectivity of effective electric field direction for tailoring the optical and electronic properties of 2D MoS 2 flake on account of its effectiveness to perturb the low dimension lattice structure. The direction-dependent electric field-induced modulation in the phonon characteristics and electronic band structure of MoS 2 has been systematically investigated based on field responsive Raman and photoluminescence measurements. The atomistic insights obtained from DFT calculations have been correlated with the experimental observations to elucidate the underlying mechanism. The applied transverse electric field is found to be significantly more efficacious than the electric field applied vertically in altering the phonon signatures and bandgap in MoS 2 , where the electrostrictive response is found to arise from the field-induced alteration in metal- chalcogen interatomic bonds. We have demonstrated a simple one-step approach to create nanostructures (such as nanoribbons and nanomesh) on MoS 2 flake of desired geometries and location by using 532 nm low power-focused laser of a Raman Confocal Microscope. We have discussed the controlling parameters for precise nanostructuring along with a detailed description of the void shape and its correlation with the crystal orientation of the plane of flake. The minimum feature size of the nonpatterns achieved in this technique is~300 nm, which is close to the diffraction limit of the laser used (532 nm). Using AFM, Raman spectroscopy and DFT modeling, an in-depth investigation has been carried out to understand the nature and mechanism of the void formation. The study shows that void always takes hexagonal or triangular shape, and the periphery of hexagonal void lies on S atoms, whereas, for the triangular void, it lies on Mo atoms of the MoS 2 crystal. This approach is more advantageous than prior reported techniques in terms of its single one-step process, easy to use, no cleanroom facilities requirement, accuracy, and controllability over designing. We have demonstrated the unperturbed capacitive behavior of MoS 2 nanostructure using Electrostatic force microscope (EFM). The comprehensive study on MoS 2nanostructures at varying tip bias voltage and lift height depicts the prominent change in phase shift at the patterned area rather than the contrast flip in-phase image of the patterned nanostructure due to the absence of free surface charges. Such phase changes at patterned nanostructure signify the capacitive interaction between tip and nanostructures at varying tip bias voltage and lift height, irrespective of their shape and size. Such capacitive response of MoS 2 nanostructures offers periodic modulation of capacitance on 2D MoS 2 flake for potential application in capacitive devices. We have explored various applications of MoS 2 nanostructure, fabricated by simple low power-focused laser irradiation techniques. Optimized geometry of these nanostructures, along with selective deposition of gold nanoparticles (AuNP), demonstrates ultrasensitive Surface-Enhanced Raman Scattering (SERS) with localized hotspots. Detailed Raman analysis shows that AuNP decorated MoS 2 nanostructure creates hotspots at the edges of the nanostructure, where enhanced Raman signal of Rhodamine B is detected. Density functional theory (DFT) calculations have been conducted to comprehend the superior deposition of AuNPs and the formation of hotspots along the artificial edges. We have demonstrated the ultrasensitive detection of RhB with SERS enhancement (~10 4) at the hotspots for RhB concentrations as low as ~10 -10 M. The AuNP decorated MoS 2 nanostructure- based SERS platform opens a new avenue to the controllable hotspots formation of desired geometry and location with high detection capability. The formation of the artificial edges on MoS 2 flake via low power-focused laser irradiation facilitates the active catalytic sites along the edges of the nanostructures for the electrochemical deposition of gold nanoparticles (AuNPs). We have demonstrated a comprehensive investigation of catalytic activities favorable for electrochemical deposition of AuNPs on created artificial edges of MoS 2 and tested its efficiency for electrochemical reduction of dinitrogen into ammonia under ambient conditions. The freshly engineered active sites on MoS 2 flake are exposed in gold chloride solution at different deposition time and potential to optimize the key factors for the superior deposition of Au. The preferentially deposited AuNPs on MoS 2 is being used as electrocatalyst for nitrogen fixation, which exhibits a high ammonia yield of 21.6×10 -8 mol s -1 cm -2 with faradaic efficiency of 4.37% at low over potential of -0.1 V as compared to prior reports. By this facile and proficient approach, active catalytic sites can be customized to desired geometry and quantity on MoS 2 flake,which paves a new perspective of engineering catalytic active sites to design potent electrocatalysts for enhancing the sensitivity in electrocatalytic reaction based on MoS 2 . In summary, this thesis demonstrates a simple, rapid, and reliable approach to fabricate the nanostructures on MoS 2 in a controlled manner of desired shape and size using a low power-focused laser etching process along with their practical applications. Our findings offer a comprehensive platform to explore wide range applications of MoS 2 based nanostructures in the field of surface plasmon resonance sensing, FET based bio-chemical sensing, opto-electronic, photonics and so on.

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