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Hybrid emitters based on two-dimensional WSe_2 and ordered plasmonic nanobumps

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Abstract. Two-dimensional transition metal dichalcogenides (TMDs), particularly tungsten diselenide (WSe_2), exhibit exceptional optoelectronic properties, including strong light-matter interactions and tunable exciton behavior, making them promising for nanophotonic applications. This work investigates a hybrid system comprising a WSe_2 monolayer integrated with a plasmonic metasurface of Au-nanobumps to enhance photoluminescence (PL) through exciton-plasmon coupling and strain-induced bandgap modulation. The WSe_2 monolayers were mechanically exfoliated and transferred onto a laser-patterned Au-nanobump array fabricated via femtosecond laser printing, offering a scalable alternative to conventional lithography. Optical characterization showed a threefold enhancement of photoluminescence intensity of WSe_2 monolayer lying on a nanobump compared to WSe_2 on a flat gold substrate, what attributed to localized plasmon-exciton interactions and strain effects. The study demonstrates a cost-effective, lithography-free approach for tailoring hybrid TMD-plasmonic systems, enabling precise control over optical properties for next-generation optoelectronic devices.

Keywords: WSe_2, emitter, nanobump, photoluminescence

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Гибридные эмиттеры на основе двумерного WSe_2 и упорядоченных плазмонных нанобампов

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Аннотация. Двумерные дихалькогениды переходных металлов (TMDs) широко известны благодаря своим уникальным оптическим и электрическим свойствам, в особенности двумерный WSe_2 обладает выдающимися люминесцентными характеристиками, что делает его перспективным кандидатом для применений в нанофотонике. В данной работе исследуется гибридная система, состоящая из монослоя WSe_2, интегрированного с плазмонной поверхностью упорядоченных Au-нанобампов, изготовленных с помощью метода фемтосекундной лазерной печати, предлагающий масштабируемую альтернативу традиционным методам литографии и травления. Оптическая характеристика показала трехкратное усиление интенсивности ФЛ монослоя WSe_2 на нанобампе по сравнению с WSe_2, лежащим на плоской золотой подложке, что можно объяснить локализованным плазмон-экситонным взаимодействием и деформационными эффектами. Исследование демонстрирует экономически эффективный подход к созданию гибридных TMDs-плазмонных систем, позволяющий точно контролировать оптические свойства двумерного WSe_2 для применения в оптоэлектронных устройствах следующего поколения.

Ключевые слова: WSe_2, эмиттер, фотолюминесценция

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Introduction

Two-dimensional materials, particularly transition metal dichalcogenides (TMDs) like MoS_2, WS_2, $MoSe_2$, and WSe_2, exhibit exceptional optical and electronic properties, including direct bandgaps in monolayer form, strong light-matter interactions, and tunable exciton behavior [1-3]. Among them, WSe_2 stands out for its superior luminescence, making it promising for optoelectronic applications. This work explores a hybrid system combining WSe_2 with plasmonic nanostructures to enhance optical properties through exciton-plasmon coupling [4]. Additionally, strain induced by transferring WSe_2 onto nanostructured surfaces modifies its bandgap and exciton transitions [5], further boosting photoluminescence efficiency.

Materials and Methods

In our work, WSe_2 monolayers were obtained using a mechanical exfoliation method and transferred to a target substrate with Au-nanobumps using a custom transfer system based on tsc2 hq graphene (HQ Graphene, Netherlands).

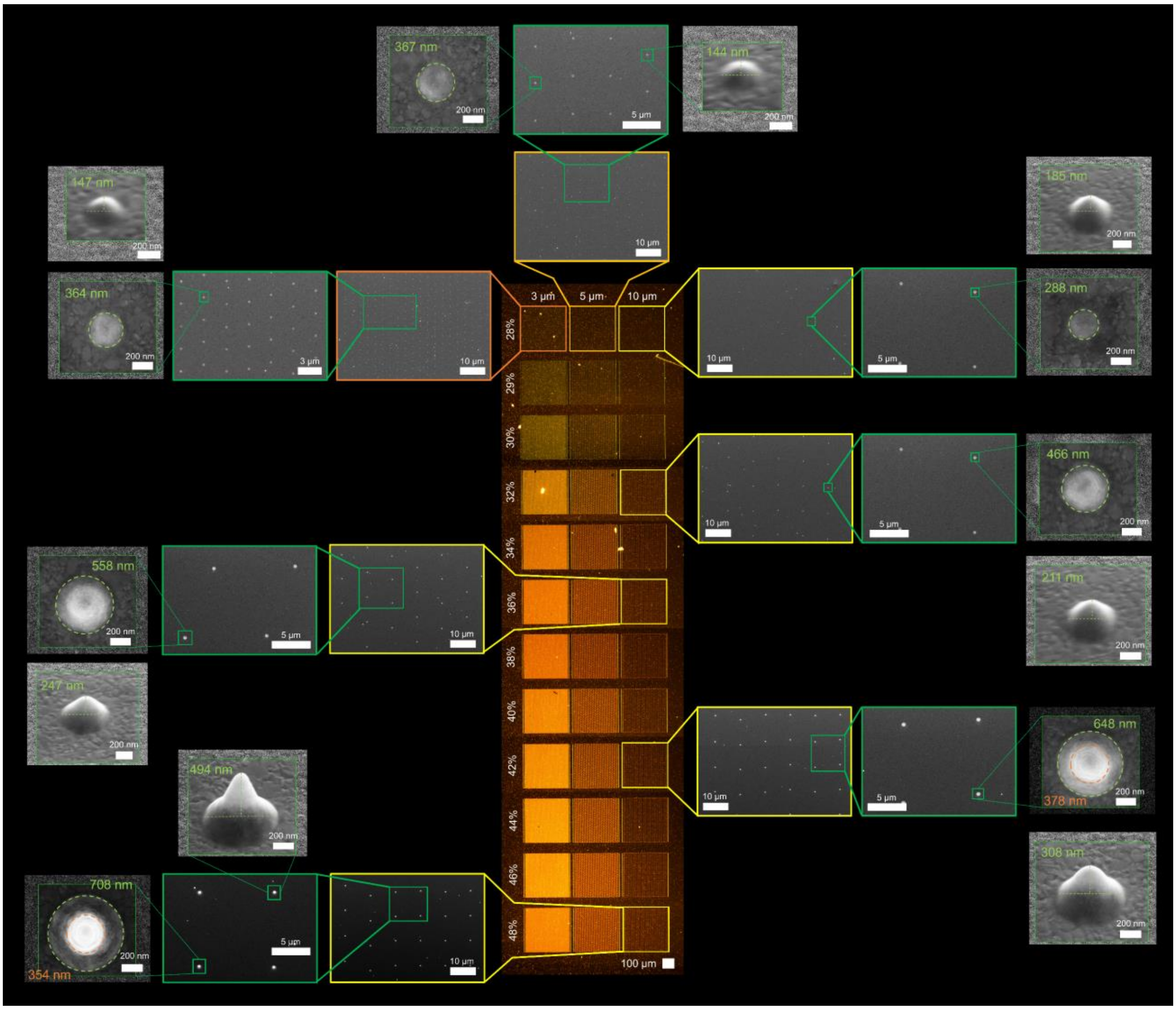

Figure 1 shows Au-nanobumps of different heights with periods of 3 μm, 5 μm and 10 μm.

Fig. 1. Au-nanobumps: optical (center) and SEM images of nanobump arrays with periods of 3, 5 and 10 μm obtained using laser pulses of different output, expressed as a percentage, where 100% corresponds to 2 nJ

The Au-nanobumps metasurface was fabricated by combination of an inexpensive and scalable magnetron sputtering of thin Au films with a direct fs-laser printing technique. First, a gold film with a thickness of 50 nm was coated over a glass substrate using magnetron deposition. Then, laser patterning of the glass-supported Au films was carried out using ≈ 200 fs second-harmonic (λ = 515 nm) pulses generated by a regeneratively amplified Yb:KGW laser system (Pharos, Light Conversion) at 50 kHz repetition rate. Output laser radiation with a Gaussian-shaped lateral in tensity profile was focused onto the Au film surface by a microscope objective (50×, Mitutoyo) with a numerical aperture NA = 0.42. A computer-driven nanopositioning platform (ANT130XY, Aerotech) ensured precise (position deviation less than 100 nm) movement of the sample surface with respect to the laser focal spot, allowing each pulse to form isolated nanobumps arranged into a square-shaped lattice with a period (Λ) defined by the sample scanning speed.

Results and Discussion

Optical properties of such a structure were characterized using Raman and photoluminescence (PL) microspectroscopy. The measurements were carried out on a Horiba LabRAM HR 800 spectrometer. The excitation radiation source was a diode-pumped 532 nm solid-state laser.

Figure 2(*a*) presents an optical image of a WSe_2 layer transferred onto a substrate patterned with an array of Au-nanobumps (147 nm height, 364 nm base diameter), fabricated using a laser pulse output of 28% (0.56 nJ), alongside the corresponding AFM image of the same region. The WSe_2 flake exhibits two distinct zones: a monolayer (1L-WSe_2) and a bilayer (2L-WSe_2). AFM analysis reveals that the flake conforms to the nanobump topography, forming a tensile "tent"-like structure over each bump, while folding between adjacent bumps is a behavior more pronounced in the bilayer regions. Disordered Au particles of varying sizes, residual from fabrication, are also observed on the substrate. PL-mapping along two bump-to-plane directions (Fig. 2(*b*)) and their spectra (Fig. 2(*c*,*d*)) demonstrate two key strain-dependent effects. First, it is an enhanced PL-intensity as the monolayer approaches the bump region (higher strain), attributed to exciton localization suppressing non-radiative recombination and plasmonic nanoantenna effects from Au-nanobumps amplifying the local electromagnetic field. Second, it is a redshift of the PL-peak (by a few nm) under tensile strain, consistent with bandgap reduction in the deformed WSe_2 lattice.

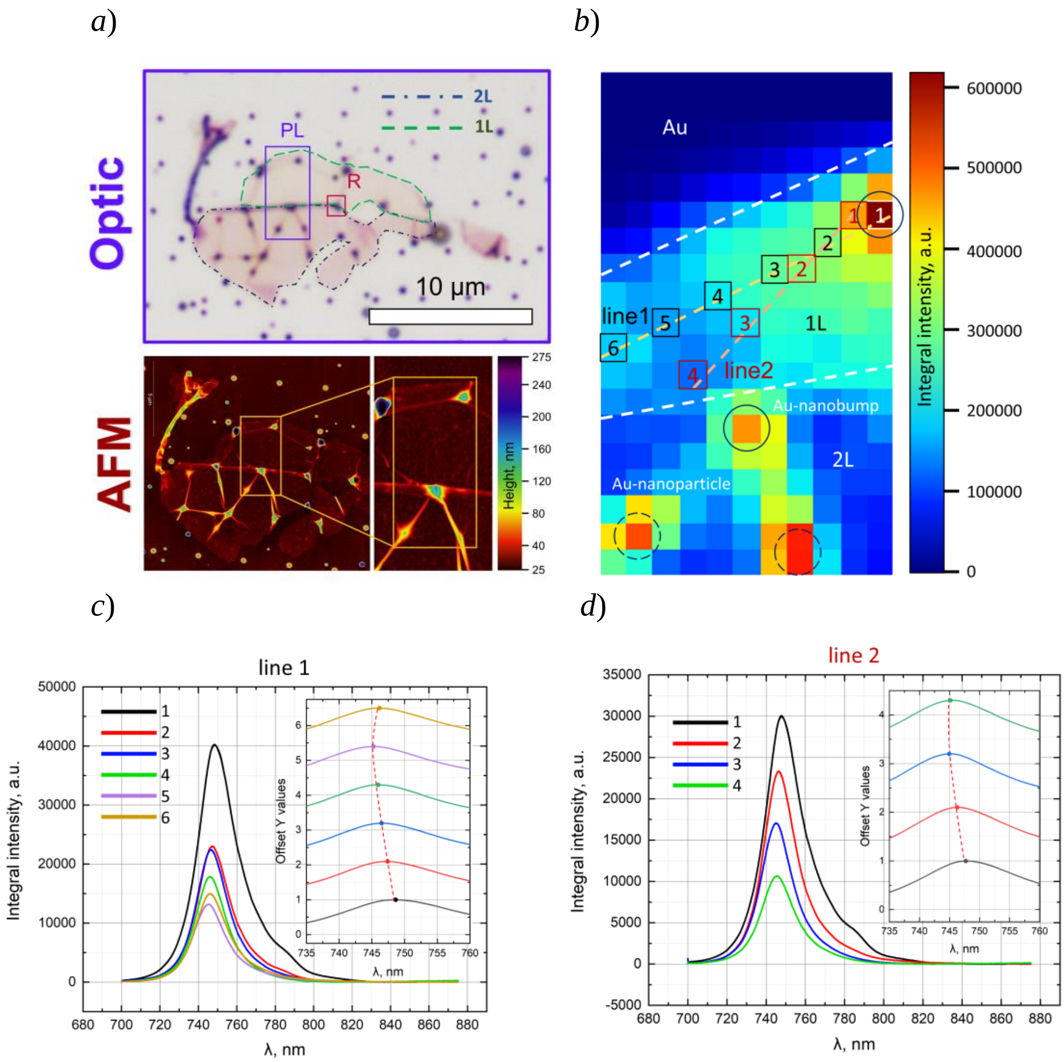

Fig. 2. WSe_2 on Au-nanobumps (28%): optical image (above) with the selected areas "*PL*" and "*R*" and AFM-image (below) (a); PL-mapping of the selected area "*PL*" representing integral intensity of a PL-spectra (*b*); PL-spectra of line 1 (*c*) and line 2(*d*) with the addition of waterfall graph showing the shift of the PL-peak respectively (*c*)

To characterize the mono- and bilayer regions of WSe_2, Raman mapping was performed in area "*R*" (Fig. 2(*a*)). The bilayer region exhibits significantly higher integrated Raman spectra intensity compared to the monolayer (Fig. 3(*a*)). The spatial boundary between 1L-WSe_2 and 2L-WSe_2 is further confirmed by the shift in the A'_1 peak position (Fig. 3(*b*)), with the bilayer showing a distinct cm^{-1}-scale shift. Representative Raman spectra (Fig. 3(*c*,*d*)) reveal key differences: the bilayer not only shows enhanced A'_1 and E' modes due to modified electron-phonon coupling but also displays new characteristic modes. The observed modes include a shear mode (ZO') at 138 cm^{-1}, arising from interlayer displacement, a layer breathing mode (LBM) at 311 cm^{-1}, indicative of strong interlayer interactions and also new optical modes at 376 cm^{-1} and 397 cm^{-1}, activated by the reduced crystal symmetry in the bilayer.

Notably, complete photoluminescence quenching was absent at the WSe_2/Au interface, suggesting high quantum efficiency in the WSe_2 monolayer. The enhanced PL intensity in this system likely stems from two synergistic effects: strain-induced modification of the WSe_2 monolayer and plasmonic enhancement from the Au-nanobumps.

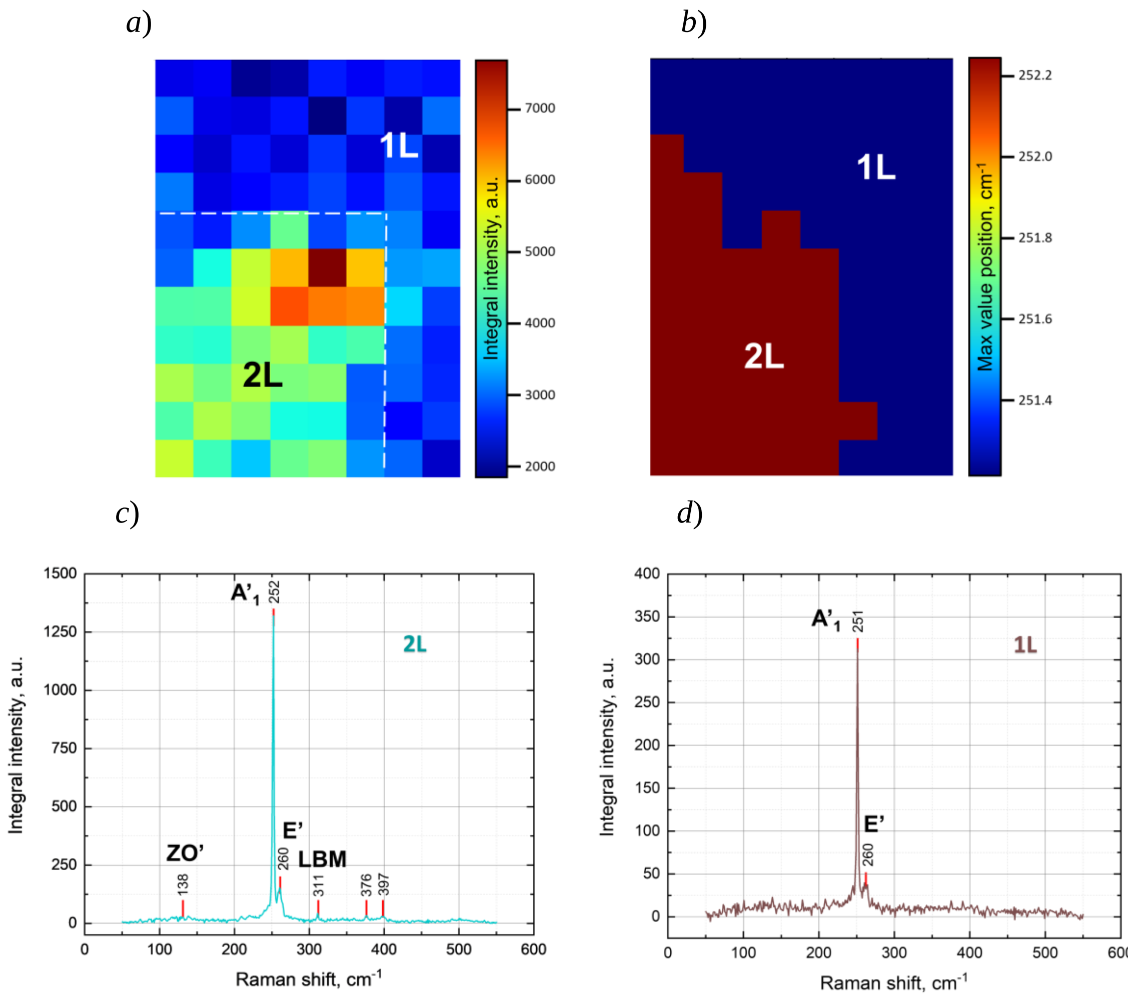

Fig. 3. WSe_2 on Au-nanobumps (28%): Raman mapping of the area R representing integral intensity (*a*) and the primary peak position in cm^{-1} (*b*) of Raman spectra; 2L-WSe2 (*c*) and 1L-WSe2 (*d*) Raman spectra with the designation of the main modes of combined light scattering

Conclusion

The innovation and practical significance of the presented work is attributed to the use of Au-nanobumps, the formation of which is carried out by a simple and effective method alternative to the traditional processes of lithography and etching. The laser radiation parameters allow controlling the geometry of the nanobumps and the lattice period, which provides the possibility of precise control over the optical characteristics of the hybrid structure. Furthermore, the ability to transfer two-dimensional WSe_2 layers onto such structures with good adhesion provides an avenue for the fabrication of efficient hybrid emitters and heterostructures based on this platform.

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