Deterministic Cost-Efficient Engineering of Quantum Light Emitters in Two-Dimensional Semiconductors

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Localized optically active quantum states in two-dimensional (2D) layered semiconductors have emerged as a promising physical system for quantum photonic technologies. Addressing the challenge of scalability and reliable deterministic generation of quantum light emitters (QLEs) at precise locations, various strategies, including strain engineering and surface treatments, have been tested. While such methods have demonstrated considerable success, their high cost renders them less accessible, highlighting the need for a more economical approach. Here, we develop a more cost-efficient and lithography-free procedure for creating large areas of spatially localized QLEs in 2D sheets. The proof-of-concept was achieved by placing the mechanically exfoliated WSe₂ and GaSe flakes over luminescent microparticles (of a specific bipyramidal geometry) dispersed onto a Si/SiO₂ substrate. Once the location of the dispersed particle on the substrate surface was identified by an optical microscope, the precise placement of the flake was achieved using all-dry viscoelastic polydimethylsiloxane stamp method.¹ The presence of several sharp corners, which are inherent to the chosen microparticle geometry, allows to create strain engineered nanoscale exciton traps. The microparticles' optical activity enables precise determination of the position of the strain induced OLEs within the flake on a (sub)micrometer scale, thereby eliminating the need for the use of additional imaging techniques. The exciton traps are optically characterized by spatially and polarization resolved microphotoluminescence as well as photon correlation spectroscopy (cf. Fig. 1). These experiments reveal the appearance of narrow elliptically polarized emission lines associated with quantum confined exciton (X) and biexciton (XX) exhibiting photon antibunching. The excellent agreement between our results and the previous reports on strain induced emission of antibunched photons in WSe_2^2 and $GaSe^3$ unequivocally confirms the suitability of our approach for the cost-effective and practical deterministic generation of highly localized QLEs. Moreover, the observed excitonic complexes suggest that these systems are suitable for the entangled photon pairs production via XX-X radiative cascade, which is still an issue in 2D semiconductor materials.

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[2] J. Kern et al., Adv. Mater. 28, 7101 (2016).

[3] W. Luo et al., ACS Photonics 10, 2530 (2023).



Fig. 1. (a) Low-temperature μ -PL spectra for different laser excitation powers on the exciton trap induced within the GaSe flake by strain deformation localized in the region above the luminescent microparticle. Inset: Optical microscopy images of GaSe flake deposited on top of the microparticle. (b) Normalized polar plots of the integrated exciton (XX, red circles) and biexciton (XX, red circles) in (a) peak intensities. (c) Evidence of photon antibunching for the X emission peak in (a).