Revealing localized excitons in WSe_2/β-Ga_2O_3

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We have investigated the optical and magneto-optical properties of monolayer (ML) WSe_2 on flakes of β-Ga_2O_3 under high magnetic fields. Remarkably, sharp emission peaks were observed and associated with localized excitons related to point defects. A detailed study of low-temperature photoluminescence (PL) and magneto-photoluminescence under high perpendicular magnetic field up to 9 T was carried out. Several sharp emission peaks have shown valley g-factors values close to -4 which is an unusual result for localized excitons in WSe_2. Furthermore, some PL peaks have shown higher g-factor values of $\approx$ -7 and $\approx$ -12 which were associated with the hybridization of strain localized dark excitons and defects. The reported results suggest that β-Ga_2O_3 is indeed a promising dielectric substrate for ML WSe_2 and also to explore fundamental physics in view of possible applications in quantum information technology.

In the last few years, there is an increasing interest in the ultra-wide bandgap semiconductor gallium oxide (Ga_2O_3) for possible applications in power electronics and UV opto-electronics[1–6]. Various growth techniques such as molecular beam epitaxy (MBE), metal organic vapor phase epitaxy (MOVPE), halide vapor phase epitaxy (HVPE), and low pressure chemical vapor deposition (LPCVD), have been developed to synthesize directly Ga_2O_3 and have resulted in high quality crystals[4,6]. Several studies were performed in the monoclinic gallium oxide (β-Ga_2O_3) because of its large breakdown field[3,5,7]. Despite of not being a van der Waals material and having highly strong ionic bonding, the β-Ga_2O_3 crystal can be mechanically cleaved and exfoliated along the (100) favorable surfaces to make ultra-thin layers which can be used in device fabrication for nanotechnology[5,7–12]. Exfoliated β-Ga_2O_3 flakes usually have thickness in the range of 150 - 300 nm[5,7–12]. However, their thickness can be further reduced by using plasma etching resulting in thickness of about 60nm[9]. The exfoliation of bulk β-Ga_2O_3 crystals and the transfer of related flakes on different materials could be an interesting strategy to prepare heterostructures and improve the quality of different based devices[5,7]. In addition, β-Ga_2O_3 has also emerged as an interesting candidate to be used as substrate[13,14] or surface protection[15] of two-dimensional (2D) materials, such as transition metal dichalcogenides(TMDs).

TMDs are well known semiconductor materials of significant interest due to their unique electronic, optical, and mechanical properties and possible applications in opto-electronics and quantum information technology[16–18]. Monolayer TMDs are direct band gap semiconductors with two inequivalent $\pm$K valleys and robust excitons[19]. Under out-of-plane magnetic fields, valley Zeeman effect and magnetic-field-induced valley polarization are observed[19,20] and these effects depend on the presence of strain, defects and doping[21–24]. ML TMDs are very sensitive to environmental ambient and usually require effective passivation and surface protection to maintain their electrical and optical performance[15,25–28]. The possibility to combine TMD with few layers of Ga_2O_3 can result in the improvement of TMD properties, enhancing device performance and enabling the development of next-generation 2D opto-electronics devices. Actually, it was shown that when few layers of crystalline Ga_2O_3 are used with TMDs, such as molybdenum disulfide (MoS_2)[13] or tungsten diselenide (WSe_2)[14,29], it resulted in excellent device properties. Furthermore, ultrathin glasses of Ga_2O_3 were also successfully used to cap ML WS_2 which resulted in an important improvement of its optical properties[15]. This integration of Ga_2O_3 with TMDs has indeed great potential for the improvement of 2D opto-electronics devices as the Ga_2O_3 can be used as a robust protective layer and passivation material for TMDs, safeguarding their electronic properties while enhancing their overall device performance[15].

Additionally, there is also an increasing interest in the generation of single photons emitters in 2D materials such as TMDs which can easily be integrated in photonic devices[16,30–36]. Single photon emitters based on monolayer (ML) TMDs are usually observed in different systems with defects (impurities or atom vacancies), wrinkles (strained regions), or using SiO_2 substrates with lithographically patterned nano-pillars to create local strain, which can induce the generation of single-photon emissions[16,30–36]. Particularly, there is also great interest to use different substrate materials to improve the optical properties of 2D materials and several possibilities to engineer and manipulate defects in these systems for possible generation of single photons emitters[36,37].

In this letter, we have performed optical and magneto-optical studies in a monolayer $WSe_2/\beta\text{-}Ga_2O_3/SiO_2$ under high magnetic fields. We observed that the Ga_2O_3 improves the optical properties of WSe_2 at low temperatures. Additionally, several sharp emissions peaks with different g-factors were also observed and associated with localized bright and dark excitons. In general, our studies provide fundamental insights on the impact of Ga_2O_3 substrates on the optical and magneto-optical properties of monolayer WSe_2/Ga_2O_3. Our results are particularly relevant for the development of devices in opto-electronics and possible applications in quantum information technology.

Fig.1 (a) shows the schematic representation of our sample. The heterostructure consists of a ML of WSe_2 on top of a flake of β-Ga_2O_3 placed on a SiO_2-Si substrate. Details of the sample preparation are presented in the supporting information file. The optical microscope image of our heterostructure is shown in Fig.1 (b). The surface morphology was investigated in detail using Atomic Force Microscopy (AFM) (see Fig.1 (c) and also Fig. S1). We found that Ga_2O_3 shows atomically flat surfaces over large areas (root mean square (rms) roughness of 1-7 nm), which is an important property to obtaining high-quality samples. Fig.1 (c) shows the AFM topography image in the region of the black square in Fig.1 (b). The Ga_2O_3 layer has a thickness of about 340 nm. The corresponding line profile (black square in Fig.1 (b)) of the β-Ga_2O_3 height is shown in Fig.1 (c). In general, our results indicate that exfoliated Ga_2O_3 crystals show good surface topography in agreement with previous results reported in the literature[7]. Fig.1 (d) shows typical PL spectra at different positions on the sample labelled S1, S2 and S3 in the optical microscope image of Fig.1 (b) using 660 nm laser excitation. We have observed several sharp PL peaks and a neutral exciton PL peak centered at around 1.75 eV (Fig. S3 and S4) with a typical full-width-at-half maximum (FWHM) of about 9.7 meV. We observed that the PL linewidth of the neutral bright free exciton is comparable to that of samples WSe_2/hBN (see Fig. S5) which suggest that Ga_2O_3 is indeed a good substrate material to improve the optical quality of TMDs. In addition, several sharp PL peaks are observed at lower energies with FWHM values in the range of 320-660 μeV at 3.6 K. These peaks seem similar to the sharp peaks observed previously in WSe_2 flakes with local strain[34,35]. Usually, the PL spectra in this system depends on the laser position due to the position dependent strain field. They are associated with localized excitons due to defects or related to hybridization of dark excitons, modulated in energy by local strain and defect states

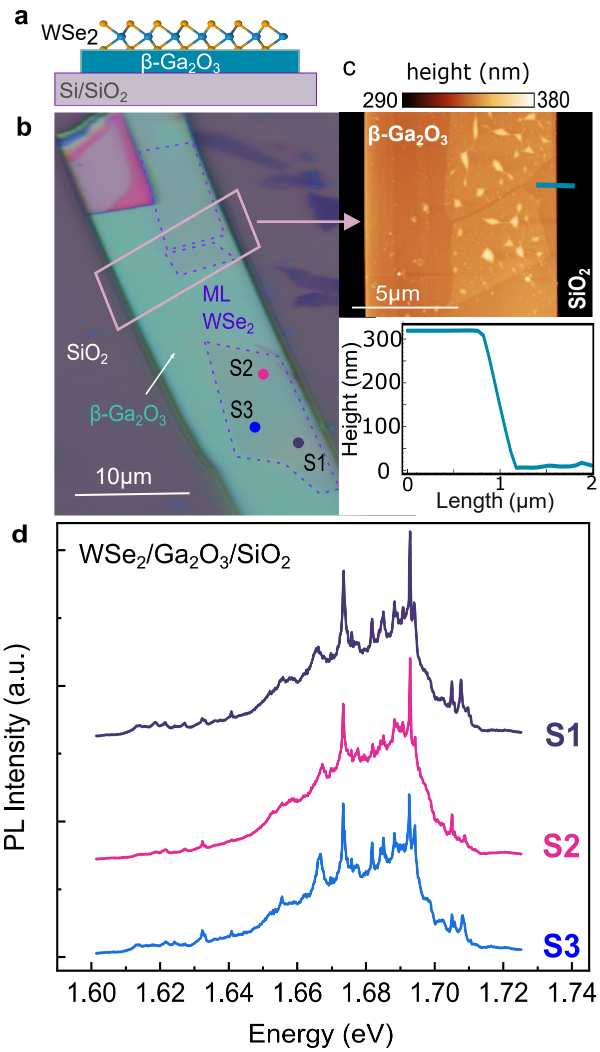

FIG. 1. (a) Schematic representation of the $WSe_2/\beta\text{-}Ga_2O_3$ heterostructure. (b) optical microscope image of the heterostructure. (c) AFM topography image in the region of the pink square in (b), with respective AFM profile line acquired (cerulean blue line). (d) typical PL spectra for different positions, labelled S1, S2 and S3 in (b), at 3.6 K. The excitation laser energy is 1.88 eV and the laser power is 50 μW.

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which are usually related to intrinsic Se vacancies[34,35,38,39]. However, we remark that for our sample (see Fig.1 (b)) several PL peaks are observed at similar PL peak energies for different laser positions on the WSe_2/Ga_2O_3 which indicates that these PL peaks could not be related to local strain effects. These peaks could be due to localized excitons by defects. On the other hand, there are also PL peaks that depend on laser position and seem related to the presence of local strain. Similar sharp PL peaks were also observed in other WSe_2/Ga_2O_3 samples (see Fig. S6). Furthermore, we have not observed any emission from the Ga_2O_3 region under visible laser excitation as expected[40,41].

A possible explanation for the type of defects in WSe_2/Ga_2O_3 samples could be related to the presence of point

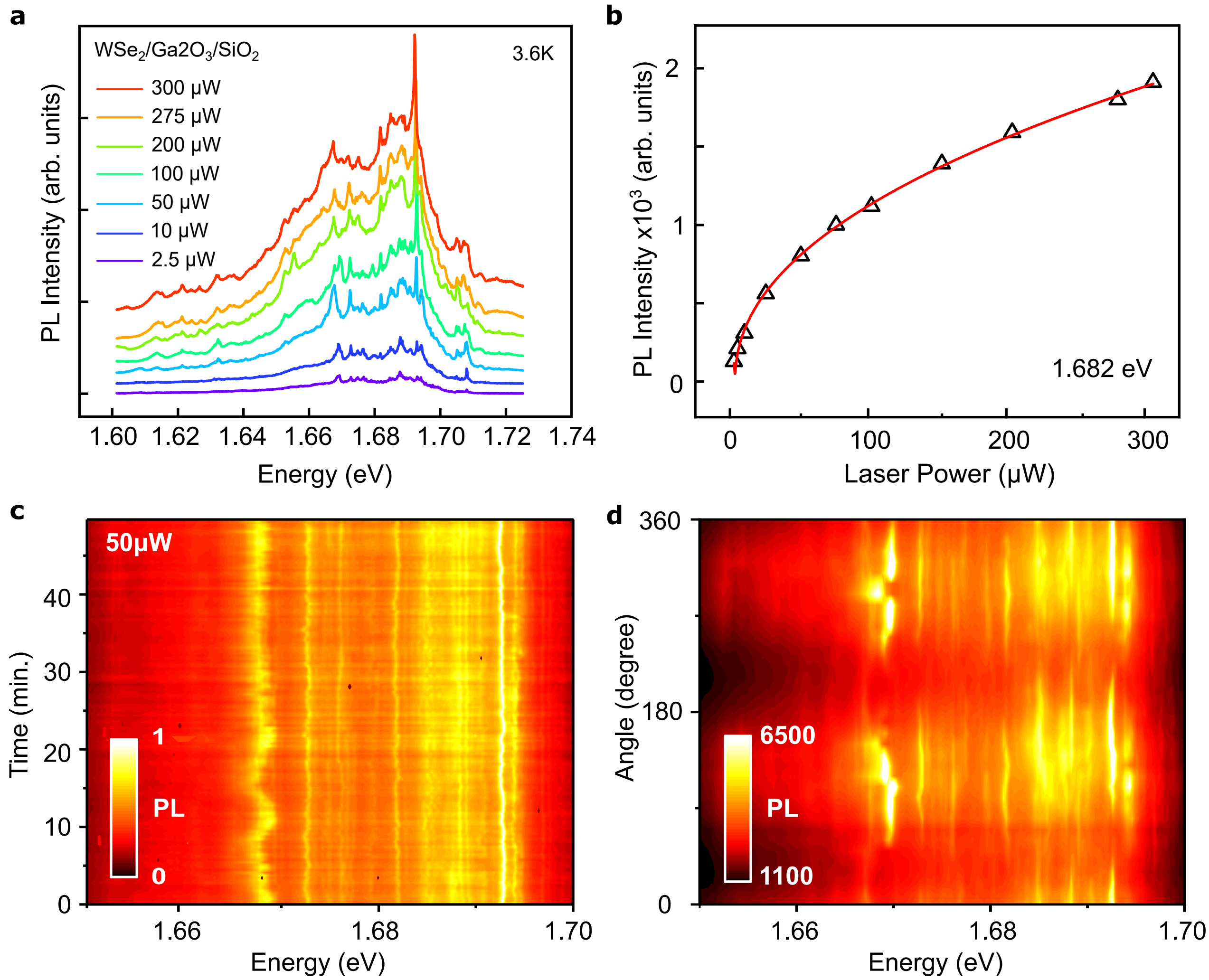

FIG. 2. (a) Laser power dependence of the PL spectra from WSe_2 (b) PL intensity as a function of laser power for the PL peak at 1.682 *eV* (c) color-coded map of the time dependence of the μ -PL emission feature in the range of 1.65-1.70 *eV* showing jittering effects (d) color-coded map of the linearly-polarized emission intensity as a function of the angle of in-plane polarization.

defects such as atom vacancies which could contribute to the localization of excitons in WSe_2[34]. However, further studies would be necessary to understand in detail the nature of these point defects and their contribution in exciton localization. In general, our PL results indicate that localized excitons in WSe_2/Ga_2O_3 have different natures.

Fig.2 (a) shows typical PL spectra of ML WSe_2/Ga_2O_3 for different laser powers at 3.6 K. We note that for all emission peaks the PL intensity increases with the increasing of laser power (lower laser powers) and saturates for higher laser powers as expected for localized emissions. Fig.2 (b) illustrates the laser power dependence of the PL intensity for a typical emission peak at 1.682 eV. We clearly observed a sub-linear behavior for lower laser intensities and a saturation with increasing laser powers as expected for localized exciton emissions. In order to investigate the stability of these emission peaks we also performed PL measurements as a function of time. Fig.2 (c) shows the color-coded map of the PL intensity as a function of time. A small jittering effect is observed and is probably related to the fluctuation of the local electric field in the vicinity of the localized excitons. Therefore, all the observed sharp peaks and also the excitonic peaks (see Fig. S7) have good stability over time. In order to verify if these optical emissions are anisotropic, we have also measured linearly polarization-resolved PL. Fig.2 (d) illustrates the color-coded map of the linearly-polarized emission intensity as a function of the angle of in-plane polarization. It is important to point out that all these sharp peaks are clearly linearly polarized in agreement of previous studies in the literature[16,30–35].

Fig.3 (a) shows the color-coded map of circularly resolved PL intensity as a function of magnetic field. The excitation laser was linearly polarized and the PL detection for positive magnetic field is σ^+ circularly polarized. The top (Fig.3 (a)) and bottom (Fig.3 (b)) panels correspond to the circularly resolved PL spectra σ^+ and σ^- for positive and negative magnetic fields, respectively. We observed a linear magnetic field dependence for the emission peaks with increasing magnetic field which is similar to previous works in the literature[35].

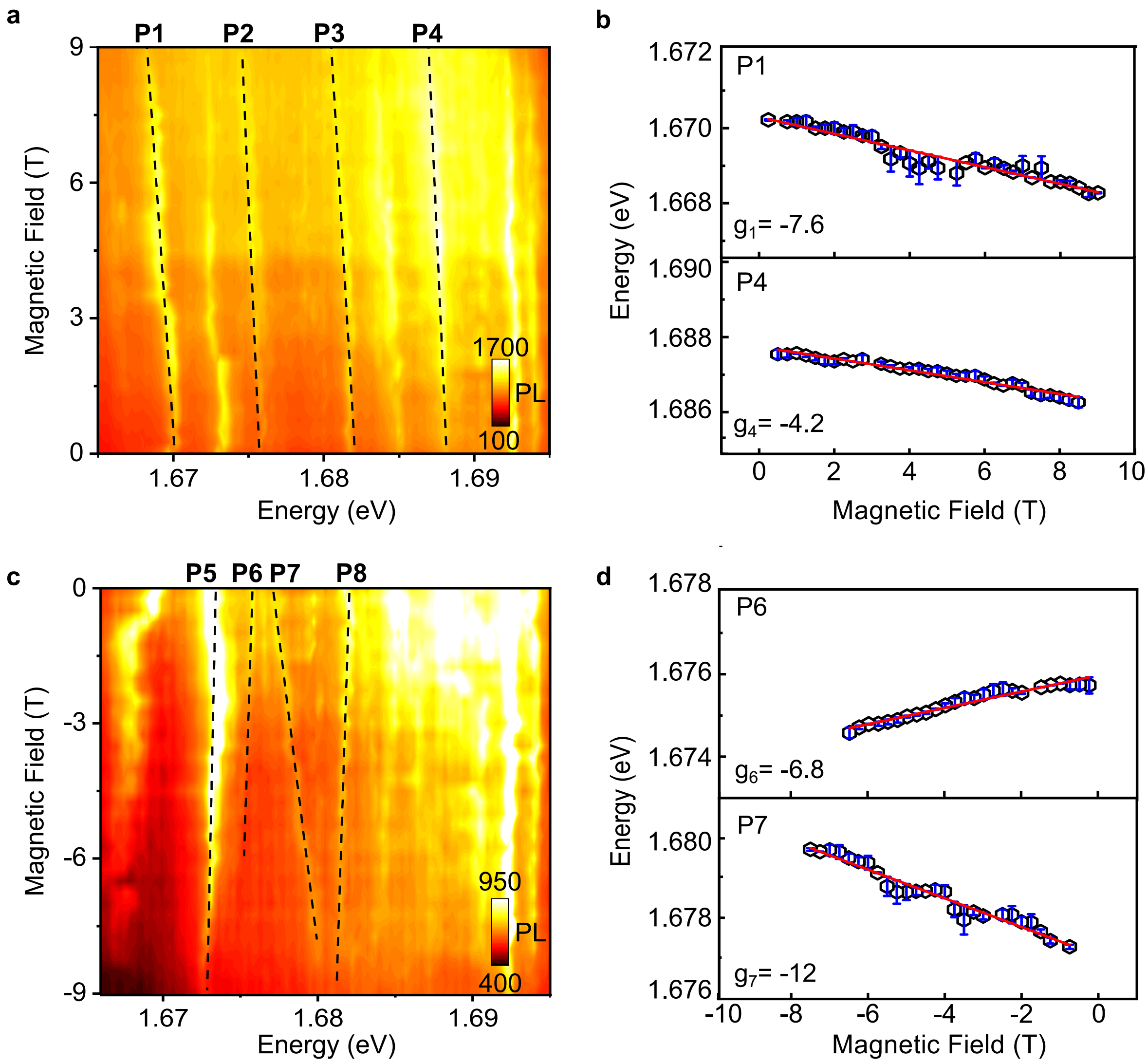

FIG. 3. (a) and (b) Color-code map of the circularly resolved μ-PL intensity as a function of positive and negative out-of-plane magnetic field respectively. These magneto-PL measurements were performed using linearly polarized laser excitation at 3.6 K. (c) and (d) energy shift versus magnetic field. The corresponding extracted g-factors are also indicated in the (c) and (d).

Furthermore, we observed that most of the emission peaks have higher PL intensity under positive magnetic field and therefore they are strongly σ^+ polarized (see Fig.S8 and Fig. S10). Few emission peaks show no important dependence with increasing magnetic field indicating different nature of localized excitons. Furthermore, Fig.3 (c) and 3 (d) show the PL peak positions of the PL peaks labelled as P1, P4, P5 and P7 in Fig.3 (a) and 3 (b) as a function of the magnetic field.

In the presence of an out plane magnetic field, the energy of each exciton state shifts as[19,42]:

$$E_i(B) = E_i(0) \pm g_i \mu_B B/2, \tag{1}$$

where μ_B is the Bohr magneton, B the magnetic field strength, i = 1,2,3... and the $\pm$ sign depends on the valley degree of freedom, associated with the circular polarization $\sigma_\pm$, and the linear term corresponds to the Zeeman shift, with a valley g-factor g_i. Here, we neglected the diamagnetic shift as they are an order of magnitude lower than the Zeeman shifts[42]. These g-factor values are directly related to the angular momenta of the valence and conduction band states involved in the exciton transition[42,43]. The extracted valley g-factors are obtained by fitting the linear curve and are shown in Fig.3 (c) and 3 (d) . Most of the extracted g-factors of the stronger emission peaks have values $\approx$ -4 (see also Fig. S11) which are similar with the values reported in the literature for free bright excitons in ML TMDs[42,43]. Furthermore, similar values were observed for localized excitons in ML $MoSe_2$[44]. This result indicates that they are probably related to localized bright excitons in WSe_2[44]. In addition, we have also measured time resolved PL (TRPL) (see Fig. S9) and which demonstrated fast decay times of $\approx$ 200 ps and seems consistent with this interpretation of localized bright excitons in WSe_2. However, the carrier dynamics could depend on the laser wavelength, and further studies would be necessary to investigate the laser wavelength dependence for the PL decay time. As mentioned above, the extracted g-factor of the neutral bright exciton in WSe_2 is also $\approx$ -4 (see Fig. S8) as expected for bright excitons. On the other hand, we remark that the magnetic field dependence of the energy shift of some peaks observed under negative magnetic field (P5, P6, P8 in Figure 3 and Figure S11) have different slopes.They could be related to doublet peaks[16,35,45,46] However, as they have very low PL intensities, it is difficult to confirm clearly the existence of these doublet peaks. Similar emission peaks were observed previously for quantum dots (QD) systems[46–48] which are related to the electron-hole exchange interaction in the presence of in-plane anisotropy that hybridizes the left-circularly σ^- and right-circularly σ^+ polarized exciton states resulting in a linearly polarized fine-structure doublet[16,45,46]. These localized PL peaks are probably due to a combination of defects, dark excitons and strain effects and are similar to the PL peaks reported previously[38,39,49] which were attributed to local strain which could be related to the presence of nano-bubbles[50]. These peaks have shown higher g-factor values of $\approx$ -7 (peaks P1 and P6) and $\approx$ -12 (peak P7) indicating the presence of different nature of localized excitons in WSe_2/Ga_2O_3. In fact, for some emission peaks we have seen two decay time components, with values of $\approx$ 232 ps and $\approx$ 1.209 ns which also evidences different nature of localized excitons. The extracted g-factors of other sharp peaks pointed out in Fig.3 are provided in the Supplementary Information (SI) (Fig. S11). In general, the comprehensive analysis of our results sheds light on the presence of different natures of localized excitons which contribute to PL spectra in the WSe_2/Ga_2O_3 heterostructure.

In conclusion, our results have shown that Ga_2O_3 improves the PL linewidth of the exciton emission in ML WSe_2/Ga_2O_3 samples. Interestingly, our results also evidenced the generation of localized emissions at lower energies in ML WSe_2 on flakes of Ga_2O_3. We have investigated in detail the magneto-PL under high perpendicular magnetic fields. We observed that several PL peaks have valley g-factors close to -4 which is an unusual result for localized excitons in ML WSe_2. These PL peaks were associated with localized bright excitons by point defects. In addition, we observed that some PL peaks have shown higher g-factor values of $\approx$ -7 and $\approx$ -12 which were associated with strain localized dark excitons hybridized with defects. These defects are probably related to point defects in WSe_2 such as Se vacancy. They may also contribute to the localization of excitons in in WSe_2. However, further research would be necessary to fully understand the nature of these defects and their relation to sharp emission peaks. Our findings suggest that heterostructures composed of exfoliated ML TMDs and flakes of Ga_2O_3 are indeed promising systems for opto-electronics and also for the generation of quantum dot-like emitters for quantum information technology.

SUPPLEMENTARY MATERIAL

Sample preparation, experimental methods and complementary PL results are available free of charge to readers.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available within the article and its supplementary material.

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