Electronic Supplementary Information

Revealing the origin of PL evolution of InSe flake induced by laser irradiation

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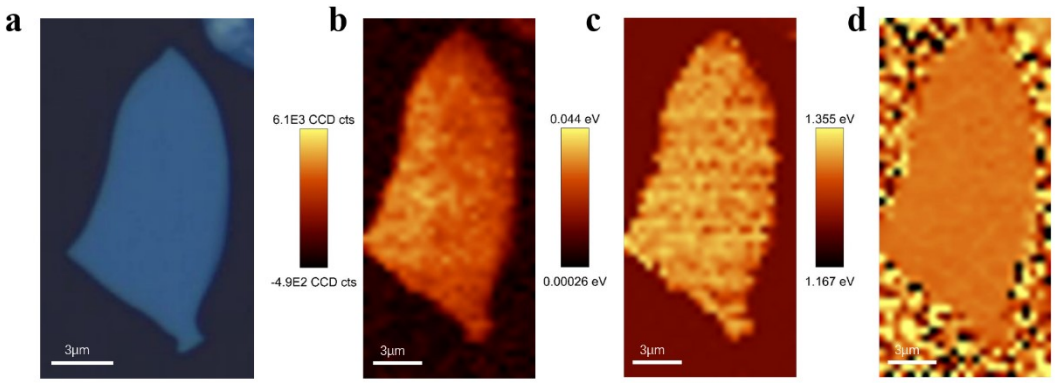

Fig S1 (a) Optical image of the InSe flake exfoliated onto a SiO2 (300 nm)/Si substrate. (b) PL mapping of InSe integrated correspondingly by intensity (c) width (d) position.

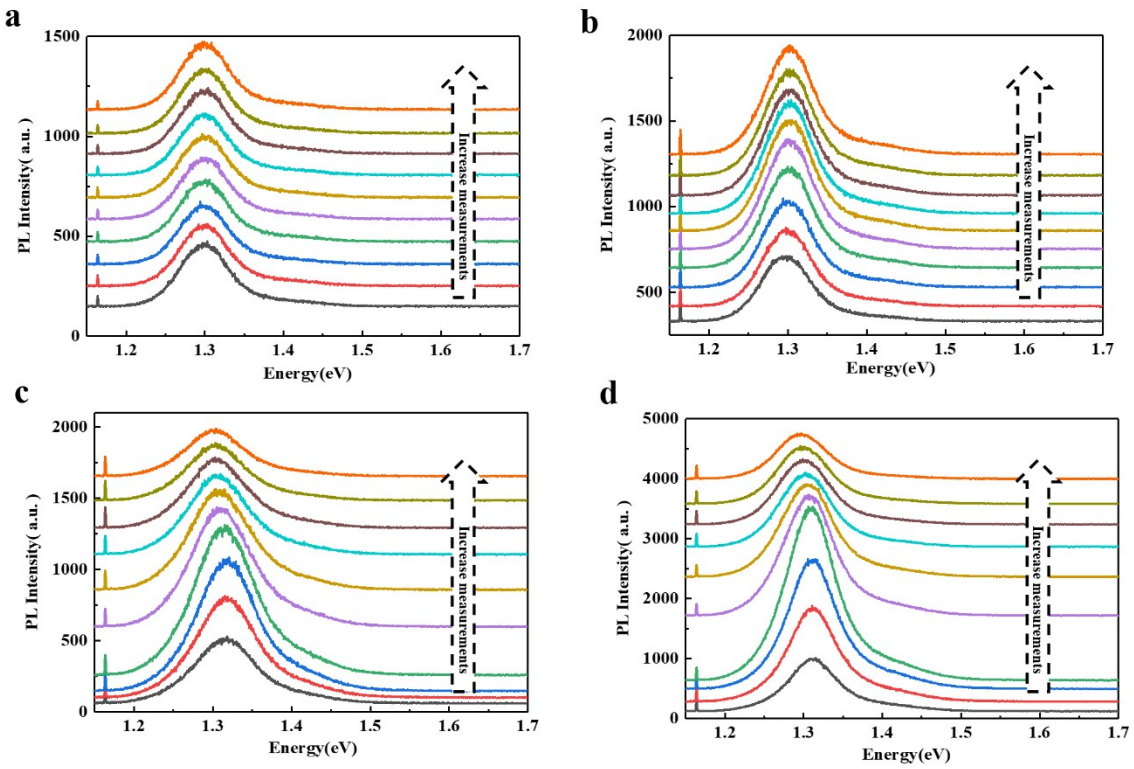

Fig S2 PL spectra of InSe flakes with increasing measurements in vacuum (a), in vacuum (after adsorption of water) (b), in air (c), in air (after adsorption of water) (d).

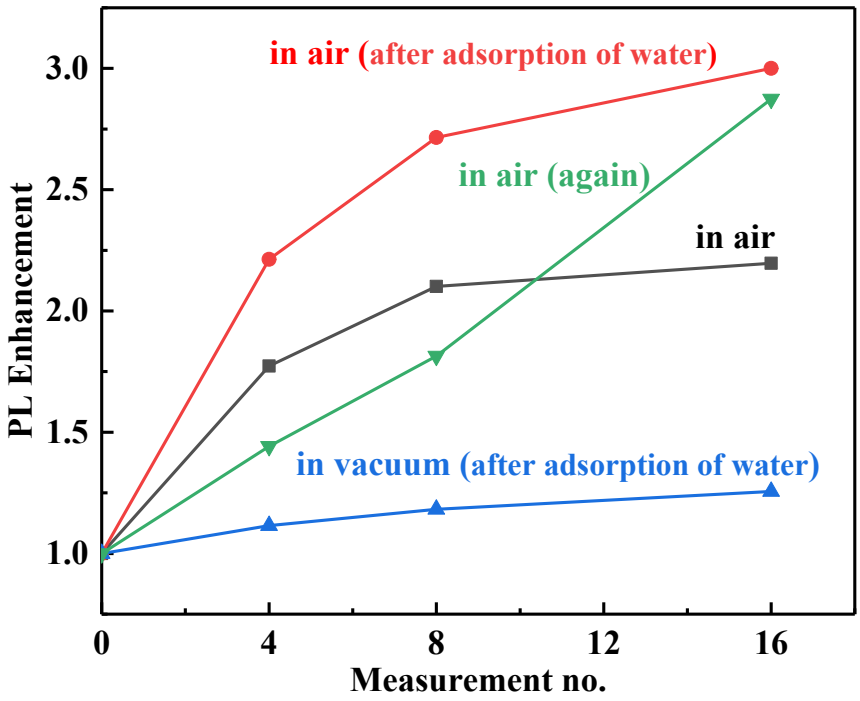

Fig S3 PL enhancement with increasing measurements in air (black); the sample in water (red); water-rich sample in vacuum (blue); water-rich sample recovers to ambient condition(green).

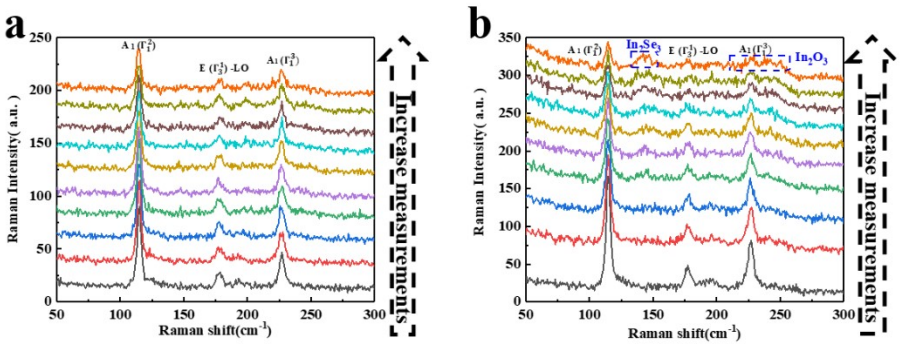

Fig S4 Raman spectra with increasing measurements under the

irradiation of 0.2mW (a) and 2mW (b) in air.

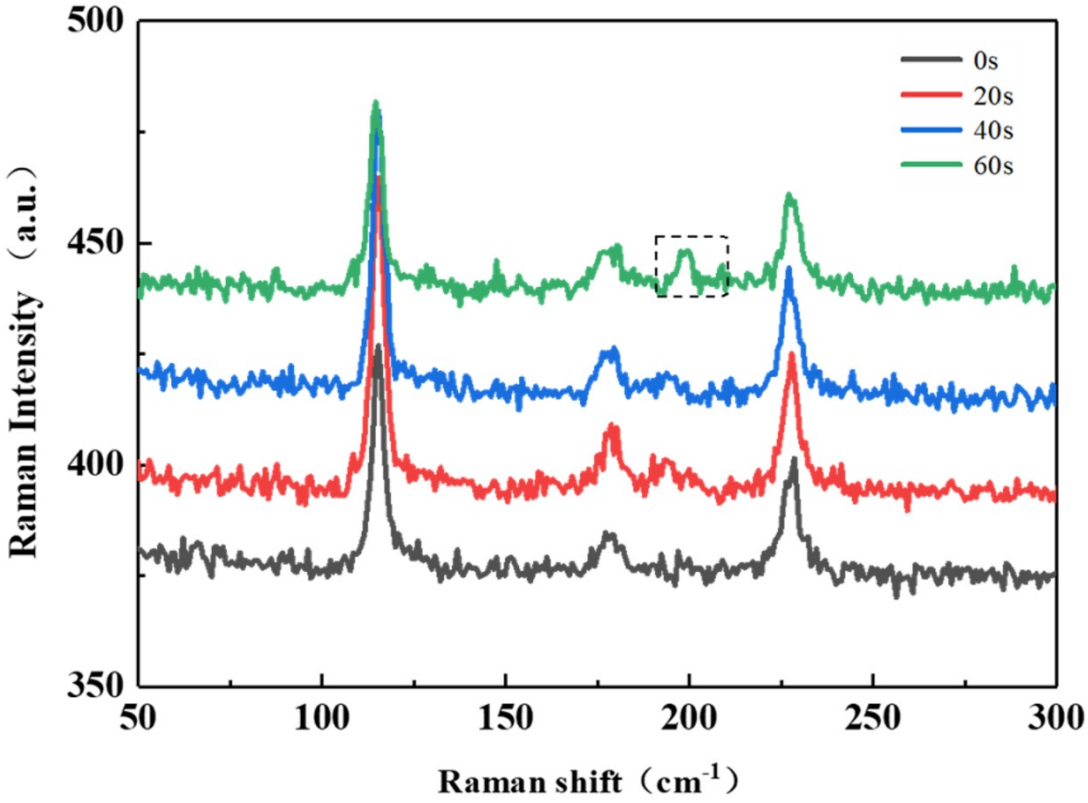

Fig S5 Raman spectra of InSe with 0s,20s,40s,60s Ar plasma irradiation durations.

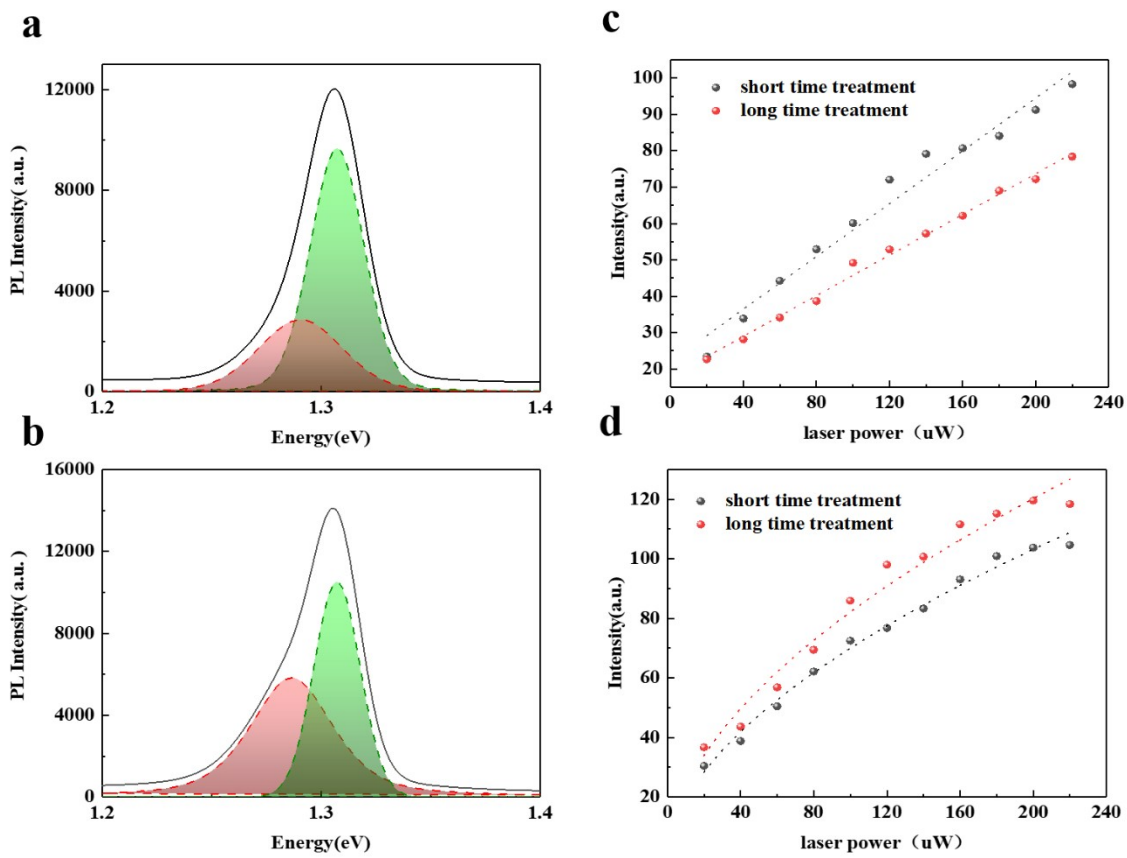

Fig S6 PL spectra at 10k with short (a) and long (b) Ar plasma treated time; PL intensity of X0 (c) and Xb (d) with different plasma treated time as a function of laser power. The solid curve is a fitting result using power law: $I\alpha P^k$.

To further confirm responsibility of introducing vacancy, we conduct the PL measurements of the plasma-treated InSe at ultralow temperature (10K) whose results are shown in FigureS6a and b. Obviously, the PL spectra consist of two peaks, we distribute the emission at ~1.28 eV and ~1.31 eV to bound excitons (Xb) and excitons (X0) by Gauss fitting, respectively[1]. The rise of the bound exciton and the decrease of exciton with increasing irradiation time of Ar plasma, consistently indicating the generation of higher density of defects as introducing vacancies[2, 3]. To quantify the defect density with different plasma irradiation, we carry out excitation power-dependent PL measurements of InSe with different treatment time and the intensities(I) are plotted as a function of excitation powers (P) in FigS6(c) (X0) and FigS6(d) (Xb). Both the intensities of Xb can be used well fitted by the power law $I\alpha P^k$ and similar k values of ~0.55 are obtained despite different times of plasma treatment, indicating that Ar^+ plasma irradiation does not change the nature of the induced defect. In comparison, the intensity of X0 increases linearly with the excitation power (FigS6c). This

nonlinear dependence means the PL intensity of bound excitons is expected to reach saturation value at high excitation power and can be interpreted as the full population of defect states with excitons at a high excitation power[2]. The saturation intensity of Xb increases with a longer plasma treatment time corresponding to a higher defect state density (Fig S6d).

REFERENCES

1. Zheng, T., et al., *Layer-number dependent and structural defect related optical properties of InSe.* Rsc Advances, 2017. **7**(87): p. 54964-54968.
2. Wu, Z.T., et al., *Defects as a factor limiting carrier mobility in WSe2: A spectroscopic investigation.* Nano Research, 2016. **9**(12): p. 3622-3631.
3. Bacher, G., et al., *INFLUENCE OF BARRIER HEIGHT ON CARRIER DYNAMICS IN STRAINED INXGA1-XAS GAAS QUANTUM-WELLS.* Physical Review B, 1991. **43**(11): p. 9312-9315.