

## Magnetic and semiconducting properties in van der Waals InSe layered crystals incorporating transition metals

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Synthesis of two-dimensional (2D) van der Waals (vdW) crystals with ferromagnetic properties is highly desirable due to their potential applications in nano-spintronics. The intensively investigated 2D crystals such as graphene and graphene-like materials, including monolayer BN sheets, silicene and layered transition metal dichalcogenides, are nonmagnetic or weakly magnetic, which limits the possibility of their use in spintronic devices. Despite the fevered interest in 2D materials in the last decade, there have been only a few theoretical [1,2] and experimental [3] reports on the synthesis and physical studies of 2D vdW crystals exhibiting magnetic properties.

Indium monoselenide (InSe) is a layered III-VI semiconductor crystal (Fig. a) with a direct band gap of 1.26 eV at 300 K, which can be tuned, due to quantum confinement, by reducing the number of layers in its nanosheets (Fig. b) [4]. In this work, we show that the magnetic properties of this vdW crystal, which is intrinsically nonmagnetic in its pristine form, can be effectively modified by electrochemical intercalation or doping with transition metals InSe:M (M = Fe, Mn, Ni, Co etc.). Bulk Bridgman-grown layered crystals of  $\gamma$ -rhombohedral InSe incorporating transition metal impurities exhibit interesting magnetic properties (Fig. c), including ferromagnetic ordering at room temperature. Most importantly, unlike most other semiconductors, InSe remains optically active even when heavily doped or intercalated with a large amount of impurities and its crystal structure is preserved (Fig. b). The InSe:M crystals can be mechanically exfoliated down to few-layer nanosheets (Fig. a) that are stable in air and optically active, as revealed by our photoluminescence (Fig. d) and Raman measurements. Thus doping or intercalation of InSe with magnetic impurities enable new functionalities, i.e. magnetic, while preserving optical and structural properties.

We report on the synthesis and characterization of InSe semiconducting vdW crystals with magnetic and electronic parameters tailored by the controlled incorporation of transition metals and/or layer thickness. The possibility of achieving highly spin-polarized ferromagnetism in heterostructure stacks (e.g. graphene/InSe:M) bound by interlayer vdW forces is discussed and explored. The unique combination of semiconducting and magnetic properties in an optically active vdW crystal does not only open new frontiers in fundamental research on semimagnetic semiconductors, but also makes this class of compounds InSe:M a promising candidate for nano-spintronic and optoelectronic applications.

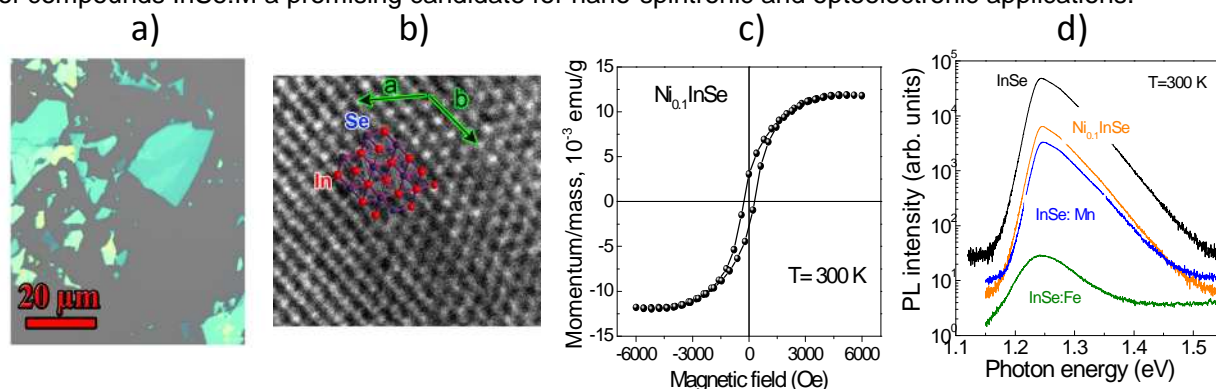


Figure. a) Optical images of InSe flakes exfoliated onto SiO<sub>2</sub>/Si. b) High resolution TEM image of InSe:Fe. c) Dependences of the specific magnetic moment on the magnetic field strength for Ni<sub>0.1</sub>InSe. d) Photoluminescence (PL) spectra of bulk InSe with and without magnetic impurities.

### References

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