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Enhanced optical, magnetic and hydrogen evolution reaction properties of $Mo_{1-x}Ni_xS_2$ nanoflakes†

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Due to exceptional electronic, optoelectronic and catalytic properties, MoS_2 has attracted extensive research interest in various applications. In the present scenario, the exploitation of noble-metal-free catalysts for hydrogen evolution is of great interest. Herein, we report the structural, optical, magnetic and electrocatalytic properties of pure and nickel-substituted MoS_2 nanostructures synthesized by the hydrothermal method. X-ray diffraction (XRD) analysis reveals that all samples exhibit the hexagonal structure of MoS_2 and the formation of NiS_2 at higher concentrations of nickel. Vibrating sample magnetometer (VSM) measurements of the $Mo_{1-x}Ni_xS_2$ nanostructures show a hysteresis loop at room temperature with a higher saturation magnetization for 1% Ni-substituted MoS_2 nanostructures, confirming the ferromagnetic behaviour of the sample. The indirect-to-direct band gap transition of few-layered nanostructures was confirmed by the optical absorption spectrum showing bands in the 600–700 nm and 350–450 nm regions. This study also highlights the excitation wavelength-dependent down- and up-conversion photoluminescence of the as-synthesized samples, providing new horizons for the design of MoS_2-based optical and spintronic devices. The electrocatalytic effect of 3% Ni-substituted MoS_2 nanostructures has been found to be higher than that of other deposit concentrations as it corresponds to the efficient hydrogen evolution reaction (HER).

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Introduction

The successful discovery of graphene by Novosolev and Geim has created a resurgence of interest in two-dimensional (2D) van der Waals crystal systems, such as boron nitride, transition metal dichalcogenides, metal oxides and hydroxides, *etc.*, for both semiconductor technology and flexible nanotechnology. Graphene, a crystalline allotrope of carbon with a 2D hexagonal crystal structure, has received significant research interest and attention due to its unique and novel electronic, optoelectronic, chemical and mechanical properties.[1,2] However, the zero-band gap energy and weak spin–orbit coupling of graphene creates a significant challenge, such as the inability to switch off graphene-based transistors, for many electronic applications. The perception of the limitations of graphene thus fueled the need of other analogues for the successful designing, fabrication and miniaturization of electronic and other flexible devices.[3] These include boron nitride, transition metal chalcogenides (TMCs), transition metal dichalcogenides (TMDs), black phosphorous, 2D metal oxides and hydroxides, *etc.* Among the family of 2D materials, TMDs have attracted significant research interest and effects in the field of material science and condensed matter physics, especially MoS_2. They are stacked into graphene-like layers with the common formula MX_2, in which M represents transition metal elements of the IV, V and VI groups, forming hexagonal layers sandwiched between two hexagonal sheets of X, where X is a chalcogen (X = S, Se, Te)[4] These can be metallic, semi-metallic or semiconducting depending on the type of metal coordinated to the chalcogen and are also affected by the insertion or intercalation of foreign materials into the MX_2 matrix. Unlike graphene, MoS_2 is a non-zero band gap energy material with large binding energy of excitons and undergoes transition from the indirect energy gap in the bulk to the direct energy gap in the few-layers. The direct optical band gap (~1.9 eV) of MoS_2 in the visible region offers a new paradigm in optoelectronic device applications.[5] Moreover, the strong spin splitting and absence of inversion symmetry in MoS_2 enable its potential applications in various nanoelectronics and spintronic devices.[6]

Dilute magnetic semiconductors (DMSs) are semiconductors where a small portion of the cation is replaced by rare earth or

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