Atomic Structure and Spectroscopy of Single Metal (Cr, V) Substitutional Dopants in Monolayer MoS_2

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Dopants in two-dimensional dichalcogenides have a significant role in affecting electronic, mechanical, and interfacial properties. Controllable doping is desired for the intentional modification of such properties to enhance performance; however, unwanted defects and impurity dopants also have a detrimental impact, as often found for chemical vapor deposition (CVD) grown films. The reliable identification, and subsequent characterization, of dopants is therefore of significant importance.
Here, we show that Cr and V impurity atoms are found in CVD grown MoS_2 monolayer 2D crystals as single atom substitutional dopants in place of Mo. We attribute these impurities to trace elements present in the MoO_3 CVD precursor. Simultaneous annular dark field scanning transmission electron microscopy (ADF-STEM) and electron energy loss spectroscopy (EELS) is used to map the location of metal atom substitutions of Cr and V in MoS_2 monolayers with single atom precision. The Cr and V are stable under electron irradiation at 60 to 80 kV, when incorporated into line defects, and when heated to elevated temperatures. The combined ADF-STEM and EELS differentiates these Cr and V dopants from other similar contrast defect structures, such as 2S self-interstitials at the Mo site, preventing misidentification. Density functional theory calculations reveal that the presence of Cr or V causes changes to the density of states, indicating doping of the MoS_2 material. These transferred impurities could help explain the presence of trapped charges in CVD prepared MoS_2.

References

[1] Alex W. Robertson *et al*., ACS Nano, 11 (2016) 10227–10236

Figures

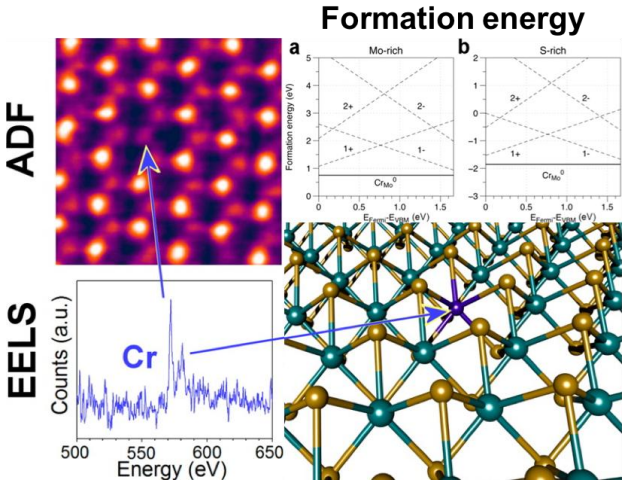

Figure 1: ADF-STEM image, EELS signal, and atomic model of Cr substitutional dopant at Mo site. DFT calculation results of the formation energy of Cr dopant is also presented.