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N- and P-doping of Transition Metal Dichalcogenide (TMD) using Artificially Designed DNA with Lanthanide and Metal Ions

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Transition metal dichalcogenides (TMDs) with a two-dimensional layered structure have been considered highly promising materials for next-generation flexible, wearable, stretchable and transparent devices due to their unique physical, electrical and optical properties. Recent studies on TMD devices have focused on developing a suitable doping technique because precise control of the threshold voltage (V_{TH}) and the number of tightly-bound trions are required to achieve high performance electronic and optoelectronic devices, respectively. In particular, it is critical to develop an ultra-low level doping technique for the proper design and optimization of TMD-based devices because high level doping (about 10^{12} cm^{-2}) causes TMD to act as a near-metallic layer. However, it is difficult to apply an ion implantation technique to TMD materials due to crystal damage that occurs during the implantation process. Although safe doping techniques have recently been developed, most of the previous TMD doping techniques presented very high doping levels of ~10^{12} cm^{-2}. Recently, low-level n- and p-doping of TMD materials was achieved using cesium carbonate (Cs_2CO_3), octadecyltrichlorosilane (OTS), and M-DNA, but further studies are needed to reduce the doping level down to an intrinsic level.

Here, we propose a novel DNA-based doping method on MoS_2 and WSe_2 films, which enables ultra-low n- and p-doping control and allows for proper adjustments in device performance. This is achieved by selecting and/or combining different types of divalent metal and trivalent lanthanide (Ln) ions on DNA nanostructures. The available n-doping range ($\triangle$n) on the MoS_2 by Ln-DNA (DNA functionalized by trivalent Ln ions) is between 6×10^9 cm^{-2} and 2.6×10^{10} cm^{-2}, which is even lower than that provided by pristine DNA (~6.4×10^{10} cm^{-2}). The p-doping change ($\triangle$p) on WSe_2 by Ln-DNA is adjusted between -1.0×10^{10} cm^{-2} and -2.4×10^{10} cm^{-2}. In the case of Co-DNA (DNA functionalized by both divalent metal and trivalent Ln ions) doping where Eu^{3+} or Gd^{3+} ions were incorporated, a light p-doping phenomenon is observed on MoS_2 and WSe_2 (respectively, negative $\triangle$n below -9×10^9 cm^{-2} and positive $\triangle$p above 1.4×10^{10} cm^{-2}) because the added Cu^{2+} ions probably reduce the strength of negative charges in Ln-DNA. However, a light n-doping phenomenon (positive $\triangle$n above 10^{10} cm^{-2} and negative $\triangle$p below -1.1×10^{10} cm-2) occurs in the TMD devices doped by Co-DNA with Tb^{3+} or Er^{3+} ions. A significant (factor of ~5) increase in field-effect mobility is also observed on the MoS_2 and WSe_2 devices, which are, respectively, doped by Tb^{3+}-based Co-DNA (n-doping) and Gd^{3+}-based Co-DNA (p-doping), due to the reduction of effective electron and hole barrier heights after the doping. In terms of optoelectronic device performance (photoresponsivity and detectivity), the Tb^{3+} or Er^{3+}-Co-DNA (n-doping) and the Eu^{3+} or Gd^{3+}-Co-DNA (p-doping) improve the MoS_2 and WSe_2 photodetectors, respectively.

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