Mutual Exchange Growth of Zintl $Eu_3In_2As_4$ and $Eu_5In_2As_6$ Nanowires by Molecular Beam Epitaxy

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Eu-based Zintl compounds have emerged as magnetic topological materials due to their antiferromagnetic (AFM) order and strong spin-orbit coupling [1]. To harness these properties and realize quantum devices, atomic-scale precision and extremely low defect densities in crystal growth are a prerequisite. We report the successful synthesis of Zintl-phase $Eu_3In_2As_4$ and $Eu_5In_2As_6$ nanowires (NWs) using molecular beam epitaxy (MBE) through a mutual cation exchange mechanism [2]. Starting with wurtzite (WZ) InAs and zincblende (ZB) InAsSb NWs grown on InAs substrates, we introduced Eu and As flux during MBE growth, which facilitated the exchange of In atoms with Eu atoms within the InAs lattice. This process resulted in the formation of single-crystallite $Eu_3In_2As_4$ and $Eu_5In_2As_6$ along the core NWs. High-resolution transmission electron microscopy (HRTEM) and energy-dispersive X-ray spectroscopy (EDS) analyses provided information on their atomic coordination and composition, confirming the structural transformation within the NWs (Figure 1). The morphology of $Eu_3In_2As_4$ grains originates from two distinctive orientational relationships between the orthorhombic $Eu_3In_2As_4$ and the underlying InAs. In contrast, $Eu_5In_2As_6$ grains extend in four directions corresponding to the tetrahedral faces of the ZB core, exhibiting an anisotropic growth preference along the *c*-axis of their orthorhombic crystal structure. Both $Eu_3In_2As_4$ and $Eu_5In_2As_6$ NWs exhibited AFM ordering, as evidenced by magnetic susceptibility measurements such as a magnetic property measurement system (MPMS) and a scanning superconducting quantum interference device (SQUID). The ability to convert InAs NWs into different Zintl phases by selecting the crystal structure of core demonstrates the versatility of this mutual exchange growth method. This study contributes to the fundamental understanding of topotactic reactions in nanostructures and highlights the potential of MBE in fabricating complex nanomaterials with precise control over composition and structure.

References

[1] Zhao, Y. *et al.* Hybrid-order topology in unconventional magnets of Eu-based Zintl compounds with surface dependent quantum geometry. *Phys. Rev. B* 110, 205111 (2024).

[2] Song, M.S., *et al.* Topotaxial mutual-exchange growth of magnetic Zintl $Eu_3In_2As_4$ nanowires with axion insulator classification. *Nature Nanotechnology* 1–8 (2024).

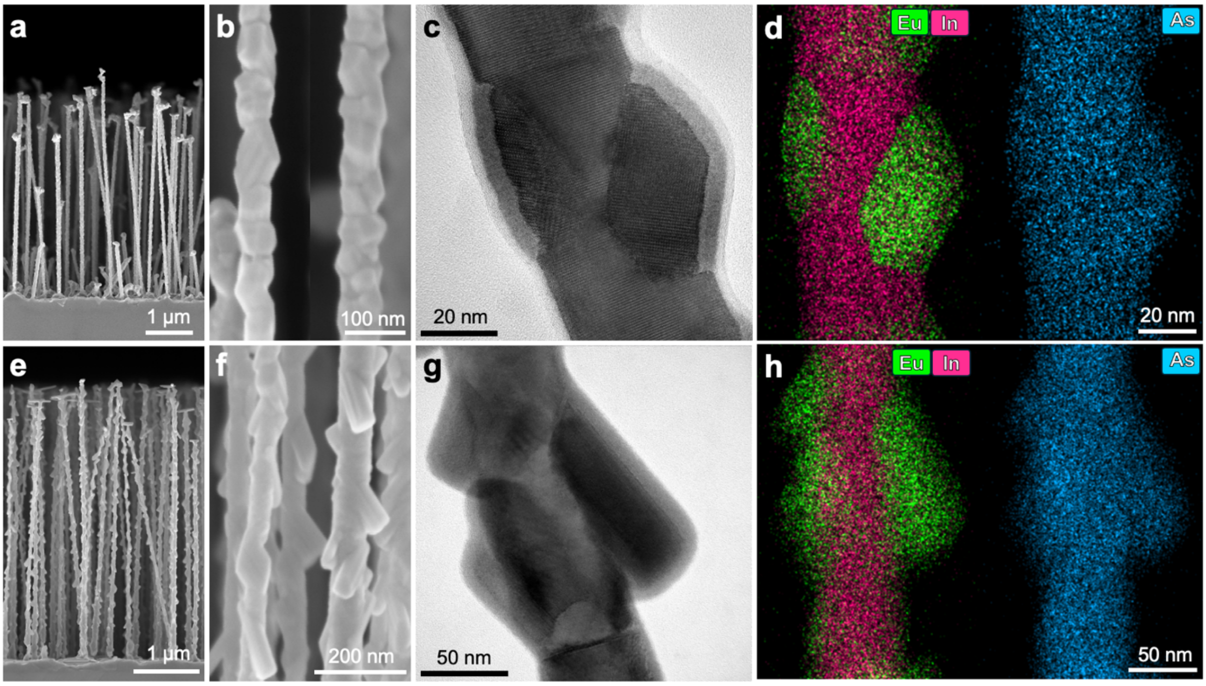

Fig. 1: (a) SEM image of as-grown $Eu_3In_2As_4$ along WZ InAs NWs. (b) The unique facets characterize the Zintl-phase $Eu_3In_2As_4$. (c) TEM image of $Eu_3In_2As_4$ grains on a WZ InAs core. (d) EDS elemental maps of Eu, In, and As (green, magenta, and blue respectively), corresponding to the area in (c). The map shows Eu and In distributed on opposite sides of the core boundary, while As is uniformly distributed throughout the core and shell. (e) SEM image of as-grown $Eu_5In_2As_6$ along ZB InAsSb NWs. (f) The Zintl-phase $Eu_5In_2As_6$ exhibits anisotropic growth direction preferences. (g) TEM image of $Eu_5In_2As_6$ grains on a ZB InAsSb core. (h) EDS elemental maps of Eu, In, and As (green, magenta, and blue respectively), corresponding to the area in (g). As in (d), Eu and In are located on opposite sides of the core boundary, with As uniformly distributed across both core and shell.