Self-scrolling in polar van der Waals nanomaterials

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Research on van der Waals (vdW) materials has advanced remarkably in the last decade, driven by the unique properties of vdW forces and emerging nanotechnologies. These advances, such as atomic-scale stacking and twisting of materials, enable the fine-tuning of electronic and topological properties in nanomaterials.[1,2] Among these breakthroughs, the concept of vdW nanoscrolls—quasi one-dimensional (1D) open structures formed by the rolling up of two-dimensional (2D) nanosheets—has paved new possibilities for dimensional engineering, providing a platform for exotic quantum phenomena and applications in nanodevices (Figure 1A).[3]

The transformation of nanosheets into nanoscrolls provides a cutting-edge approach to fabricating dimension-engineerable nanomaterials and offers distinct advantages, such as enhanced light-matter interactions, tunable chirality, and improved mechanical stability. This dimensional reduction not only preserves the intrinsic quantum confinement of the 2D nanosheets, but also introduces new functionalities, including curvature and symmetry breaking, which are unattainable in flat geometries. Creating these nanoscrolls has been challenging due to the need for precise control over parameters such as layer number, curvature, and chirality, as well as the inherent difficulty of synthesizing high-quality, high-yield nanoscrolls in a scalable, reproducible manner.

The formation of these nanoscrolls is fundamentally driven by the competition between bending energy and vdW forces. Theoretical studies suggest that spontaneous 2D sheet scrolling occurs when the gain in free energy from vdW interactions outweighs the elastic bending energy induced by the rolling process (Figure 1B). For certain materials, misfit layers or janus sheets can naturally roll into nanoscrolls driven by lattice mismatch, providing a

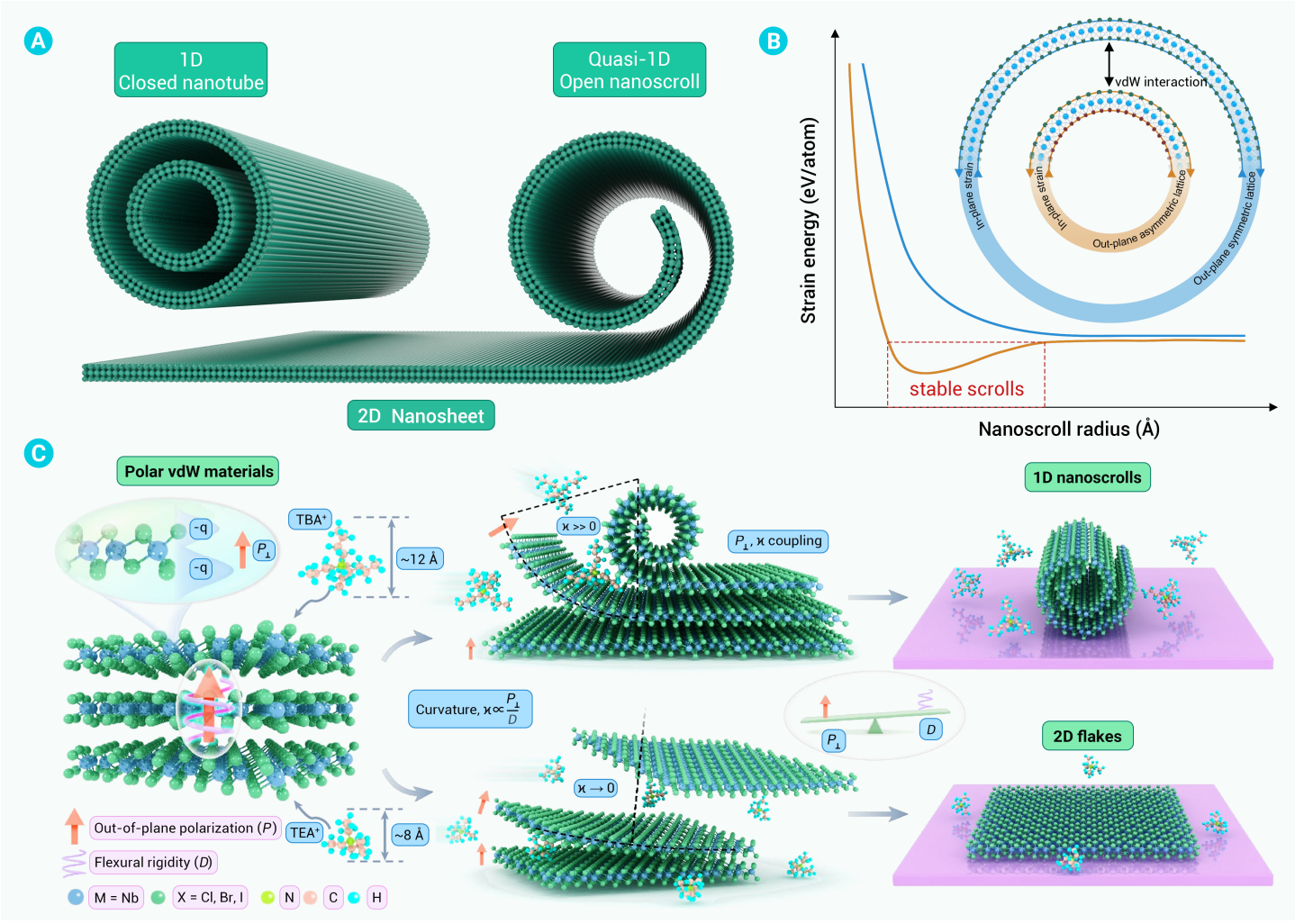

Figure 1. Schematic illustration of dimensional engineering and self-scrolling design in polar van der Waals nanomaterials (A) Structural relationship between one-dimensional closed nanotubes, two-dimensional flat nanosheets, and quasi-one-dimensional open nanoscrolls. (B) Energy and structural insights into the bending behavior of nanostructures in van der Waals materials. (C) Schematic diagram of the controlled synthesis of nanoscrolls and nanosheets from layered polar materials in M_3X_8 and M_3QX_7 (M = Nb, Ta; X = Cl, Br, I; Q = S, Se, Te), along with their alloy derivatives.[4]

template for understanding the behavior of other vdW materials. Despite decades of extensive research in nanotechnology and chemical engineering, conventional methods for nanoscroll fabrication remain reliant on external driving forces such as capillary forces, template-assisted strategies, and electrostatic interactions. While these methods have been successful in creating artificial nanoscrolls, they often suffer from limitations in scalability, uniformity, and control over critical structural parameters. The lack of a natural driving force for rolling has hindered the systematic exploration of nanoscrolls and their potential applications in energy storage, ion transport, and nonlinear optics.

Zejun Li et al. presents a groundbreaking solution: polarization-driven spontaneous scrolling.[4] This innovative method harnesses the intrinsic out-of-plane electric polarization ($P_{\perp}$) of polar vdW materials to drive the formation of high-quality nanoscrolls with nearly 100% yield during electrochemical intercalation (Figure 1C). The unique advantage of this technique lies in its use of internal polarization as the sole driving force, thereby overcoming the limitations of externally driven methods. By carefully selecting intercalant molecules of varying sizes, researchers can modulate the polarization strength, triggering spontaneous scrolling when the polarization surpasses the material's bending stiffness. This approach enables scalable, solution-based fabrication of high-quality nanoscrolls, expanding the potential for using polar van der Waals materials in next-generation nanodevices. Specifically, when large molecules such as TBA^+ are intercalated, they induce substantial edge curvature in polar vdW materials. This curvature enhances the intrinsic polarization, enabling it to surpass the critical threshold and overcome the material's bending stiffness, thereby triggering spontaneous scrolling. In contrast, smaller molecules like TEA^+ induce minor edge bending, producing insufficient polarization enhancement to overcome this critical limit and resulting in conventional 2D nanosheets.[5] This out-of-plane polarization-driven spontaneous scrolling strategy holds promise for extension to a broader class of polar materials, thereby markedly advancing research on 1D nanoscroll structures.

Nanoscrolls fabricated by this polarization-driving strategy exhibit exceptional optoelectronic properties. Their curved architecture breaks inversion symmetry, facilitating second-harmonic generation with approximately 100-fold higher SHG intensity than 2D flakes, achieving performance levels rivaling those of state-of-the-art 2D nonlinear optical materials such as $NbOI_2$ and $NbOCl_2$. Structurally, these nanoscrolls exhibit well-defined crystallographic orientation preferences, scrolling exclusively along two mutually perpendicular directions [100] and [$\bar{1}$20], thereby exhibiting well-defined chiral characteristics. Building on this foundation, the fabricated photodetectors achieve high-efficiency broadband circularly polarized light detection, demonstrating considerable promise for chiral optoelectronics. Furthermore, the flexibility of the electrochemical intercalation strategy enables the construction of multifunctional heterostructures. They developed an electrochemical co-intercalation of metal ion strategy that successfully produced 10 types of interlayer metal-ion-embedded hybrid nanoscrolls. While preserving the fundamental characteristics of 1D nanostructures, this approach introduces novel physical functionalities through ion doping (such as spin-curvature coupling), thereby providing new material platforms for designing spintronic devices and quantum components, thereby bridging the gap between fundamental nanomaterials and cutting-edge functional devices.

Looking ahead, this polarization-driven scrolling technique promises to expand the library of materials suitable for vdW nanoscrolls. Multifunctional nanomaterials can be discovered and explored more quickly using a combination of theoretical simulations and high-throughput screening. Exploring the interplay between curvature and exotic physical properties, such as topological states and superconductivity, reveals the unique electronic states of nanoscroll materials and opens new avenues for the design of flexible quantum devices. The realization of large-scale manufacturing of 1D nanoscrolls provides a robust foundation for the development and industrial application of nanodevices. These advancements position nanoscrolls at the forefront of research in quantum light sources, chiral sensing, and spintronic devices, providing a robust platform for future breakthroughs in quantum information technologies.

The discovery of polarization-driven spontaneous scrolling in polar vdW materials represents a revolutionary step in the field, with enormous potential for fabricating 2D materials into quasi-1D nanoscroll open structures with highly controllable, scalable, and intrinsic mechanisms for producing high-quality nanoscrolls, which holds immense promise for future technological advancements. This innovation tackles the two main problems of low yield and structural instability in nanoscroll fabrication, while also opening up new opportunities to exploit nanoscrolls' unique properties (such as hollow architecture and the precisely tunable coaxial vdW gaps of nanoscrolls create highly ordered nanoconductors) across a wide range of applications, from energy storage to quantum computing. Additionally, the self-encapsulated structure effectively mitigates the susceptibility of 2D materials' surfaces to environmental disturbances, thereby protecting their structural and chemical stability. Meanwhile, the nanoscroll structure provides a protected space for functional modification and spatially confined coupling, making nanoscrolls a versatile and stable platform for studying multifunctional 2D materials. This strategy establishes a new paradigm for modulating structures and exploring optoelectronic functions in low-dimensional materials.

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DECLARATION OF INTERESTS

The authors declare no competing interests.