

# Towards all-epitaxial van der Waals magnetic tunnel junctions

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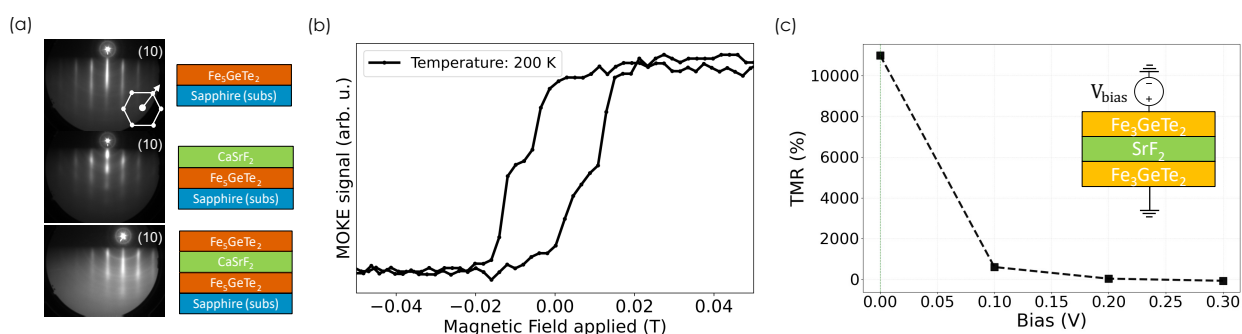
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The newly discovered class of 2D magnetic materials paves the way for the realization of ultracompact spintronic devices with enhanced functionalities for memory and logic applications. Among the different 2D magnets discovered so far,  $\text{Fe}_n\text{GeTe}_2$  ( $n=3,4,5$ ) compounds have caught special attention due to their itinerant ferromagnetism and high Curie temperatures approaching room temperature<sup>1</sup>. Density Functional Theory (DFT) studies have shown that the small overlap between majority and minority spin states across the 1<sup>st</sup> Brillouin zone leads to predictions of extremely large tunneling magnetoresistance (TMR), making these van der Waals magnets ideal for 2D magnetic tunnel junctions (MTJs)<sup>2</sup>. However, the choice of the insulating barrier is critical, as its  $k_{\parallel}$ -resolved spin filtering can significantly affect the MTJ performances<sup>3</sup>. In this context, we investigate alkaline earth fluorides ( $\text{CaF}_2$ ,  $\text{SrF}_2$ ,  $\text{BaF}_2$ ), a class of insulators characterized by F-terminated inert surfaces that form van der Waals-like interfaces with 2D materials<sup>4</sup>. Previous attempts to realize  $\text{Fe}_n\text{GeTe}_2$  ( $n = 3,4,5$ )-based MTJs relied on mechanical exfoliation<sup>5</sup>, yielding nanoflakes with limited control over thickness and lateral dimensions. Here, we employ molecular beam epitaxy (MBE) to grow 2D heterostructures on  $\text{cm}^2$  areas with precise control over chemical composition and thickness. We demonstrate the growth of fully epitaxial van der Waals-MTJs with either  $\text{Fe}_3\text{GeTe}_2$  or  $\text{Fe}_5\text{GeTe}_2$  electrodes and a  $(\text{Ca,Sr})\text{F}_2$  tunnel barrier (Fig. 1a)<sup>6</sup>. By employing electron and X-ray diffraction, atomic force microscopy, high-resolution electron microscopy, magneto-optical Kerr effect (MOKE) and X-ray magnetic circular dichroism, we assess the structural and magnetic properties of the heterostructures (Fig. 1a,b) and demonstrate atomically sharp and weakly interacting  $\text{Fe}_n\text{GeTe}_2$ /fluoride interfaces. We further performed *ab initio* DFT calculations that establish  $\text{SrF}_2$ -like fluorides as very promising tunnel barriers for providing extremely large TMR values when combined with  $\text{Fe}_n\text{GeTe}_2$  (Fig. 1c).

## References

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## Figures



**Figure 1:** (a) RHEED patterns demonstrating the epitaxial growth of an  $\text{Fe}_5\text{GeTe}_2/(\text{Ca,Sr})\text{F}_2/\text{Fe}_5\text{GeTe}_2$  MTJ. (b) MOKE measurement at 200 K, showing different coercivities of the two  $\text{Fe}_5\text{GeTe}_2$  electrodes. (c) TMR as a function of applied bias for a  $\text{Fe}_3\text{GeTe}_2/\text{SrF}_2/\text{Fe}_3\text{GeTe}_2$  MTJ, from DFT *ab initio* calculations.