

Zinc-blende half-metallic ferromagnets are rarely stabilized by coherent epitaxy

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for an actual substrate material that has a natural lattice constant close to a_s^0 . Furthermore, examination of the curvature of $E_{\text{epi}}^{\text{ZB}}(a_s)$ can tell us if this phase is mechanically stable under axial c/a distortions²⁷ or even if it could be dynamically unstable.²⁸ Such calculations have to be performed both for ferromagnetic (FM) and antiferromagnetic (AFM) spin arrangement, examining if epitaxial stability comes with the desired FM form of magnetism. The main questions hence are: Is $E_{\text{epi}}(a_s)$ vs a_s bound? If it is, does $D_{\text{epi}}(a_s)$ cross zero (i.e., does epi-ZB ever become lower energy than epi-NiAs)? If it does, is epi-ZB ferromagnetic? If it does, is there a ZB substrate material whose natural lattice constant is close to a_s^0 (deciding the thickness²⁴ that can be grown)?

We have carried out such first-principles calculations for a number of binary systems that are thought^{10–20} to be potential FM half-metallic material in their ZB form, and which were previously predicted to have rather small D_{bulk} values, spurring hope that they can be grown pseudomorphically.

TABLE I. Calculated magnetic ground states (F=ferromagnetic, A=antiferromagnetic) of bulk and epitaxial configuration of the zinc-blende- (ZB) and NiAs-type (NA) structures of various transition-metal binaries. We also give the calculated equilibrium lattice constants a and c/a ratio of the bulk phases. D_{bulk} denotes the amount by which the bulk NiAs structure is stabler than the bulk ZB structure at their respective equilibrium volumes (Fig. 1d). We indicate whether or not the epitaxial ZB structure becomes more stable than the epitaxial NiAs structure at some critical substrate lattice constant a_s^0 , and the lattice-constant range where epitaxial ZB is half metallic (HF).

	NiAs-type		ZB-type		ZB HM range			
	Bulk a Å, c/a	Epi	Bulk a Å	Epi	D_{bulk} (eV)	Epi NiAs → Epi ZB? a_s^0 Å	Bulk s Å	Epi s Å
MnAs	F, A	F	F, A	F, A				

Unfortunately, we found such hopes to be mostly unfounded because the epitaxial energy curves $E_{\text{epi}}^{\text{ZB}}(a_s)$ of the ZB form are only weakly bound, especially for MnAs, CrSb. $D_{\text{epi}}(a_s)$ does not cross zero for MnAs, CrSb, CrAs, CrTe at least up to the substrate lattice constant available from the largest lattice constant ZB materials InSb or CdTe ($a_s \approx 6.5$ Å). Thus, under epitaxial growth for a large range of a_s , the NiAs-type structure continues to be more stable than the ZB-type structure, just as is the case for bulk growth. $D_{\text{epi}}(a_s)$ does cross zero for CrS and CrSe at $a_s^0 = 5.75$ and 6.24 Å, respectively. However, having done so, the epitaxial ZB forms of CrS is AFM, not FM, so the system is not half-metallic. Finally, CrSe does satisfy all conditions and could conceivably be grown as half-metallic ZB structure for $a_s \approx 6.2$ Å (e.g., on a $\text{Cd}_x\text{Zn}_{1-x}\text{Se}$ substrate). This study shows that one could identify half-metallic epitaxial ZB structure, but that the theoretical methodology that must be used is different than hitherto practiced.^{10–20}

To perform epitaxial calculations for NiAs-on-ZB we need to find a relationship between the lattices of the film and the substrate such as those formulated in Ref. 8 for NaCl-on-ZB. To do so we inspect Fig. 2, which shows several possible lattice relationships between the NiAs-type lattice and ZB lattice. Indeed, some NiAs-type films, such as MnAs, have been^{9,29} experimentally known to grow along the $\bar{1}100$ direction, with every fourth MnAs (0002) plane matching along the (0001) direction every sixth GaAs (1120) plane. This is pattern A shown in Fig. 2. Since c/a is very close to 3/2 for all the studied NiAs-type binaries in a wide range of volumes (see Fig. 3d), we assume that these NiAs-type epitaxial films will grow as pattern A in Fig. 2, i.e., we fix c/a at 3/2, and $a_s = a_s^0 \sqrt{2}$. In general, the minima of the hydrostatic E_{bulk} vs a curves coincide with that of the epitaxial E_{epi} vs a_s curves, e.g., see the FM NiAs-type curves for MnAs and CrTe in Fig. 3. However, this is not the case when

