

due to the strong out-of-plane anisotropy we experimentally observe, this contribution would be small.

We now discuss the mechanism inducing the crossover from a pronounced AFM towards an FM order as the number n of the QLs in the MBT_n stacking sequence increases (Sec. II B and Fig. 2). Our calculations for the ordered $\text{MnBi}_6\text{Te}_{10}$ (Sec. II C) yield an – albeit small – AFM coupling. Although the increasing $K/2J$ ratio with increasing n [21] certainly helps to stabilize the FM order for $n \geq n_{\text{FM}} = 2$ in our samples (see Sec. II B and Fig. 2c), the fact that previous studies reported $n_{\text{FM}} = 3$ [22–25] hints at an additional phenomenon being involved. In sections II A and II C we have established the presence and the role of Mn/Bi antisite defects that can drive enhanced FM properties. As our numerical modelling has shown both for the MBT_0 with the strongest interlayer AFM coupling and for the MBT_1 with a periodic alternation of SLs and QLs, the motif of an intermixing pattern determines whether ferro- or antiferromagnetism is preferred. Hence, the observed magnetic properties of our $\text{MnBi}_6\text{Te}_{10}$ samples likely originate in a prevalence of intermixing patterns that favor the FM order.

In this context, a comparison between the MBT_n series and the analogous Sb-based family $(\text{MnSb}_2\text{Te}_4)(\text{Sb}_2\text{Te}_3)_n$ (MST_n) becomes relevant. The FM order is more dominant even for $n = 0$ in MST_n and MBST_n , and, as widely accepted by now, is driven by the Mn/Sb intermixing [62–65]. This phenomenon is much stronger in MST_n than in MBT_n since it is facilitated by closer atomic radii of Mn and Sb. On the other hand, the impact of the intermixing-induced FM state in MnSb_2Te_4 on its band topology is still under ongoing debate [9, 62, 63, 66–70]. Intrinsic p-type doping in MST_0 hampers clear-cut spectroscopic observations of the possible surface states and, thus, an ultimate conclusion about its topological nature. Furthermore, QAHE realizations in the MST_n have not been reported. In fact, Ref. [63] argues the importance of further studies on how intermixing impacts bulk and surface magnetism in the established topological MBT_n materials, but focuses on the MST_0 instead, because the necessary intermixings were not accessible by the bismuth analog at that time. So far intermixing in the MBT_n has been discussed mostly in the terms of its influence on the Dirac-point gap [71], while the consequences for the magnetism are quite unclear. En route to understanding the broader role of intermixing, a recent study reveals its crucial influence on the magnetic coupling in MnBi_2Te_4 [72], and our current work pinpoints the particular antisite defects that enhance (or suppress) the local FM coupling in the MBT_n series.

We have established that the FM properties of our crystals are conditioned by the underlying cation intermixing. It is instructive to examine whether this relationship holds true for the other published works. Whereas Mn deficiency in $\text{MnBi}_6\text{Te}_{10}$ is often found by x-ray spectroscopy [22, 24, 25], the related intermixing has been

scrutinized only in Ref. 23. On the one hand, their and our samples have such commonalities as the presence of Mn/Bi intermixing, the absence of cation vacancies, and a strongly mixed occupancy on the $3a$ site. On the other hand, there are also substantial differences: The mixed $3a$ occupancy is more pronounced in our sample, in which we find 56% Mn (and 44% Bi), than in the sample studied in Ref. 23, which has 83% Mn (and 17 % Bi). Most importantly, the Mn distribution over the $6c$ positions is distinctly different: We observe a higher Mn concentration in both $6c$ sites of the QLs, i.e. up to $\text{Bi}_{1.86}\text{Mn}_{0.14}\text{Te}_3$ vs. $\text{Bi}_{1.92}\text{Mn}_{0.08}\text{Te}_3$ in Ref. 23, and up to 2 % Mn in the outer positions of the SL that are reported defect-free in Ref. 23. In general, the $3a$ site in our crystals is more Mn-depleted, so that these “stray” Mn atoms, which find no space on the $3a$ site, disperse over the entire layered stack by occupying $6c$ sites. In accordance with our theoretical deliberations in Sec. II C (models S_1 to S_4), the less pronounced intermixing and the presence of swapped Mn only in one of the two $6c$ sites of the QLs in the samples of Ref. [23] accords with them featuring an AFM ground state.

The question of why intermixing takes place and which kind of defects are more likely to occur is evidently very relevant and, at the same time, a complex one. First, recent literature has shown that antisite cationic defects have the lowest formation energy and are energetically favorable to form in both MnBi_2Te_4 and MnBi_4Te_7 [73]. It has been argued that such defects provide an effective way to release a lattice strain effect which occurs within the septuple layer of MnBi_2Te_4 due to a mismatch between the MnTe and Bi_2Te_3 structure fragments. Second, as argued below, variations in the intermixing patterns of $\text{MnBi}_6\text{Te}_{10}$ samples produced by different groups may stem from subtle differences in the synthetic procedures, pointing to the relevance of finite temperature effects for the relative stability of different defects. This would be no surprise since these compounds are formed at elevated temperatures and are metastable at room temperature as we have previously shown [31, 32].

Comparing our growth conditions (see Sec. IV) for $\text{MnBi}_6\text{Te}_{10}$ to those of Refs. 23, 33, and 34 reveal differences in dwelling times, the starting and quenching temperatures, and the composition of a melt, which may well account for the various intermixing patterns. In general, we observe reproducible Mn concentrations and magnetic behavior in our MBT_2 crystals (Fig. 2 and Fig. S4) for an applied tempering profile [31, 74], suggesting that the cation intermixing is a temperature-regulated phenomenon. We do not argue that this process is fully governed by the thermodynamics, since crystallization of the MBT_n from a heterogeneous melt is strongly kinetics-driven. Yet it seems plausible, that the resulting intermixing pattern is governed by a given synthetic protocol. Strong correlations between the synthesis temperatures and the resultant cation disorder and magnetic order have been, by now, undoubtedly established at least for MnSb_2Te_4 [62, 74, 75]. Since the Bi-analogs have