



FIG. 3. Crystal- and magnetic-structure models and their DFT energies. (a) Schematics of the structure models with various Mn/Bi intermixing scenarios.  $S_0$  is the defect-free case while the panels from  $S_1$  to  $S_{15}$  visualize the structural differences to  $S_0$ . Models  $S_1$  to  $S_4$ : antisite Mn (green) is in the 6c site of the QLs; models  $S_5$  to  $S_8$ : antisite Mn is in the 6c site of the SLs; models  $S_9$  to  $S_{15}$ : antisite Mn is in the 6c positions of both QLs and SLs. (b) Schematics of the constructed magnetic arrangements. Each model has a distinct order between the Mn magnetic moments in the 3a and 6c positions which are conditioned by the respective structure model in (a). Ferromagnetic order with an out-of-plane orientation of the moments is assumed within each atomic layer. (c) Total energy difference between an antiferromagnetic ordering model (1, 2 or 3) of each structure model ( $S_0$  to  $S_{15}$ ) and the respective fully ferromagnetic configuration, as obtained from the scalar-relativistic DFT calculations. The dashed line follows the ground state energy, zero corresponding to the FM phase.

gory antisite Mn occupy the 6c positions of *only* the SLs ( $S_5$  to  $S_8$ ). In the third category, the Mn atoms are distributed over the 6c positions of *both* the QLs and the SLs ( $S_9$  to  $S_{15}$ ).

For each structure model, we consider four possible magnetic arrangements: a fully spin-polarized FM order and three different AFM models sketched in Fig. 3b. They all have in common that the Mn moments order FM within any given atomic layer, but vary in the magnetic couplings between the adjacent atomic layers along the stack. In the AFM<sub>1</sub> and AFM<sub>2</sub> models, the Mn spins in the two consequent 3a positions are oppositely coupled. The coupling between the 3a site and all intermixed Mn neighbours in the 6c site(s) is either AFM (AFM<sub>1</sub>) or FM (AFM<sub>2</sub>), respectively. The AFM<sub>3</sub> model realises parallel spin arrangement in the 3a sites, while they couple AFM with the Mn defects in all 6c positions.

Fig. 3c discerns what is an energetically favorable magnetic arrangement for each considered structure model of  $\text{MnBi}_4\text{Te}_7$  as compared to the fully spin-polarized FM state. For a given model, if any AFM model obeys  $E_{AFM} - E_{FM} < 0$ , we conclude that antiferromagnetism

is preferred. On the other hand, if all AFM models fulfill  $E_{AFM} - E_{FM} > 0$ , we define the fully FM phase as the ground state.

A clear trend in the magnetic order as a function of the underlying Mn/Bi intermixing pattern can be established. All but one models of the first two categories, where the intermixed Mn cations occupy *either* the SLs *or* the QLs, show an AFM configuration as the lower energy state. This preference reverts markedly if the antisite Mn distributes over the 6c positions of *both* the QLs and the SLs: five out of the seven constructed structure configurations of this category prefer the FM phase. A closer look reveals that the preference for the FM state is particularly prominent in those structure models ( $S_{12}$  to  $S_{15}$ ), in which the Mn cations occupy the nearby 6c positions not separated by the 3a positions – namely, when continuous magnetic exchange pathways exist between the antisite Mn ions.

These results establish a strong correlation between the magnetic structure and the Mn distribution along the stacking direction. When an antisite Mn is located only in one of the two 6c positions of the QLs – a situation ex-