F. XMCD Sum Rule and Peak Asymmetry Analysis

The MLFT analysis yields the *local* magnetic moments based on the spectral shape. The sum rules, in turn, relate the integrated Mn $L_{2,3}$ XMCD and x-ray absorption spectral intensity to the long-range ordered orbital and spin magnetic moments near the surface [49–51] (sec. IV and sec. S.V-A). We show their application to data for sample #4 in Fig. 6. After a background correction (Fig. 6a), we obtain the XAS and XMCD data (Fig. 6b), from which we calculate the integrals p, q and r required for the sum rule analysis (Fig. 6c). Finally, in Fig. 6d we show the distribution of the resulting values for m_{XM}^{spin} obtained by applying the analysis 16384 times while randomly varying the sum rule parameters within reasonable error margins. Also taking into account some ambiguity in the choice of the XAS background due to the rather featureless but intense tails of the preceding Te $M_{4,5}$ edges allows us to estimate the errors (sec. S.V). We obtain $m_{\rm XM}^{\rm spin} = 2.3 \pm 0.25, \ m_{\rm XM}^{\rm orb} = 0.1 \pm 0.15$ and $m_{\rm XM}^{\rm tot} = 2.4 \pm 0.3$ (in $\mu_{\rm B}/{\rm Mn}$, table II). The same analysis for sample #1 yields $m_{\rm XM}^{\rm tot} = (2.2 \pm 0.35) \mu_{\rm B}/{\rm Mn}$, which is compatible with sample #4 within the error.

An alternative way to obtain $m_{\rm XM}^{\rm spin}$ is to analyze the XMCD L_3 peak asymmetry [52–54], which avoids the problems arising from uncertainty in p due to the overlap of the L_3 and L_2 peaks (sec. S.V-B). We obtain $m_{\rm XM}^{\rm spin} = (2.55 \pm 0.25) \mu_{\rm B}/{\rm Mn}$, which is about 10% larger than the sum rule result.

Table II also shows that the orbital moment is negligible within the error, as expected for a predominant d^5 configuration (sec. II E). The total moment $m_{\rm XM}^{\rm tot}$ obtained with surface sensitive XMCD is reduced by about 40% in comparison to the one obtained with bulk sensitive SQUID magnetometry. Increasing the field to 6 T brings the moment $m_{\rm XM}^{\rm tot}$ to $3.9\,\mu_{\rm B}$, i.e. closer to $m_{\rm SQ}^{\rm tot} = 4.2\,\mu_{\rm B}$ and to the theoretical maximal moment (sec. II E).

It is important to keep in mind that the indicated errors of the XMCD results only take into account statistical fitting and background estimate effects. However, the shallow probing depth can further bias the outcome (sec. IID and ref. [55]): It is reasonable to expect that the QLs and the outer (6c) positions of the SLs might contain slightly canted Mn, as well as a few percent of paramagnetic and possibly even AFM (with respect to the 3a positions) Mn, see sec. IIA, sec. III and Fig. 1. Already for SL surface termination, the FM ordered Mn sheet (3a positions) is buried about $0.55 \,\mathrm{nm}$ below the surface. At the same time, there is Mn in 6c positions closer to the surface, both in the very same SL and in QLs, which can terminate the surface in different parts of the sample. Therefore, already for a mean probing depth (MPD) of about 1 nm, the contribution of the FM Mn would be notably suppressed. MPD values between 1 and 2.5 have been reported for this photon energy range [56, 57]. Given the presence of the heavy elements Te

Table II. Comparison of the magnetic moments obtained from analysis of surface sensitive XAS and XMCD data measured at $T \approx 3.5$ K in a 0.15 T field with bulk-sensitive SQUID magnetometry results at T = 2 K and the same field (in units of $\mu_{\rm B}/{\rm Mn}$). The error bars are explained in Supporting Information sec V.

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Sum rules:	$m_{ m XM}^{ m spin}$	2.3 ± 0.25	
	$m_{ m XM}^{ m orb}$	0.1 ± 0.15	
	$m_{ m XM}^{ m tot}$	2.4 ± 0.30	
Asymmetry:	$m_{ m XM}^{ m spin}$	2.55 ± 0.25	
SQUID:	$m_{ m SQ}^{ m tot}$	4.2 ± 0.2	

and Bi, which might even further attenuate the escaping electrons, an MPD close to 1 nm is not unrealistic and the involvement of probing depth effects is well conceivable.

III. DISCUSSION

The major finding of our study is that the surface of MnBi₆Te₁₀ exhibits FM properties comparable to its bulk, with a robust FM subsystem in the topmost septuple layer, which can interact with the topological surface states. Indeed, recent ARPES reports suggest the opening of an exchange gap of about 15 meV [58].

As outlined in sec. IIF, probing-depth effects can at least partially explain the 40% reduction of the XMCDderived remanent moment as compared to the SQUIDderived bulk moment. Additional surface effects might influence magnetism and therefore warrant consideration. First of all, the incomplete out-of-plane coordination by magnetic neighbors of the topmost SL suppresses the out-of-plane magnetic interactions and interrupts the exchange paths of the antisite Mn ions. However, since these interlayer interactions are weak (sec. IIC), additional theoretical scrutiny would be required to elucidate, what role their further suppression might play. Second, a competition between the demagnetizing field and the crystalline anisotropy might result in a canting and a suppression of the XMCD signal – an effect which must, however, be small due to our finding of a strong out-of-plane anisotropy, see sec. IID and the inset of Fig. 4a. Third, a combined study involving DFT and XMCD suggests that TSS couple to magnetic atoms such as Co and Mn at the surface of Bi₂Te₃, contributing to their interaction by a RKKY-like mechanism [59]: Due to their highly localized nature, electrons in the TSS interact more strongly with magnetic moments than electrons in the bulk. This might contribute to the differences between the magnetic properties at the surface and in the bulk. Our results encourage similar calculations for MnBi₆Te₁₀. Finally, the interaction with the TSS might also result in a slight helical canting away from the out-of-plane orientation, driven by Dzyaloshinskii–Moriya interactions [60, 61]. Again,