

Spin dynamics in the antiferromagnetic phases of the Dirac metals $AMnBi_2$ ($A = Sr, Ca$)

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The square Bi layers in $AMnBi_2$ ($A = Sr, Ca$) host Dirac fermions which coexist with antiferromagnetic order on the Mn sublattice below $T_N = 290\text{ K}$ (Sr) and 270 K (Ca). We have measured the spin-wave dispersion in these materials by triple-axis neutron spectroscopy. The spectra show pronounced spin gaps of $10.2(2)\text{ meV}$ (Sr) and $8.3(8)\text{ meV}$ (Ca) and extend to a maximum energy transfer of $61\text{--}63\text{ meV}$. The observed spectra can be accurately reproduced by linear spin-wave theory from an Heisenberg effective spin Hamiltonian. Detailed global fits of the full magnon dispersion are used to determine the in-plane and inter-layer exchange parameters as well as on the magnetocrystalline anisotropy constant. To within experimental error we find no evidence that the magnetic dynamics are influenced by the Dirac fermions.

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I. INTRODUCTION

Following the observation of the topological properties of electrons on the honeycomb layers of graphene, the universal characteristics of massless dispersing low-energy quasiparticles have been realized across a variety of condensed matter systems. The ternary bismuthides $AMnBi_2$ ($A = Ca, Sr$)^{1–4} are a recent addition to this family of so-called Dirac materials. The Bi square layers of $AMnBi_2$ have been found to show the same unusual transport characteristics as graphene or topological insulators^{5–8}. Due to the suppression of backscattering processes, the electronic and thermal conductivity are enhanced, and the large separation of Landau levels produces a large linear magnetoresistance. Indeed, angle-resolved photoemission spectroscopy (ARPES) has provided direct evidence of the linear band crossings in both $SrMnBi_2$ and $CaMnBi_2$ ^{9,10}, with a highly anisotropic Dirac cone.

Among other Dirac materials, these bismuthides attract special interest because their Dirac fermions may couple to transition-metal states, promising an indirect experimental handle to tune the topological bands. Below $T_N^{Sr} \approx 290\text{ K}$ and $T_N^{Ca} \approx 270\text{ K}$, the large divalent Mn ($3d^5, S = 5/2$) magnetic moments of magnitude $\approx 3.7\mu_B$ in these materials align parallel to the c -axis and form antiferromagnetic structures¹¹. The two compounds were found to differ in the sign of their interlayer coupling, resulting in ferro- and antiferromagnetic stacking of Néel-ordered layers in $CaMnBi_2$ and $SrMnBi_2$, respectively¹¹. An interpretation based on first principles calculations suggests that in the ferromagnetically stacked case ($CaMnBi_2$), the Dirac bands may provide an itinerant inter-layer exchange path and thus directly couple to the magnetic ground state¹¹. This appeared to

be supported by a weak resistivity anomaly observed at T_N in $CaMnBi_2$, but not in $SrMnBi_2$ ¹¹. Earlier transport studies, however, had not registered such an anomaly in either $SrMnBi_2$ (Ref. 4) or $CaMnBi_2$ (Refs. 12 and 13).

In metallic magnets a coupling between the ordered magnetic moments and conduction electron states can reveal itself in the magnetic excitation spectrum. For example, there can be damping due to spin-wave decay into the Stoner continuum, anomalies in the magnon dispersion due to modifications of the exchange interactions by conduction electron states, or gap formation due to an additional Kondo energy scale.

Here we report on a single-crystal neutron inelastic scattering study of $SrMnBi_2$ and $CaMnBi_2$ in the magnetically ordered state. Our analysis shows that the magnon spectrum in both materials can be accurately reproduced from a Heisenberg model describing a local-moment, quasi-two-dimensional (2D) antiferromagnet. The model includes nearest- and next-nearest-neighbor in-plane exchange interactions and a weak inter-layer exchange interaction, together with an easy-axis anisotropy. We did not find any anomalies that would suggest significant coupling between the magnons and conduction electron states. The interlayer coupling is smaller than found in the reference compound $BaMn_2Bi_2$, consistent with the larger separation of the Mn spins along the c -axis in $AMnBi_2$.

II. EXPERIMENTAL DETAILS

The preparation and characterisation of the single crystals used in the experiments has been reported previously¹¹. Polycrystalline $AMnBi_2$ was first synthesized by solid-state reaction of the elements. Single

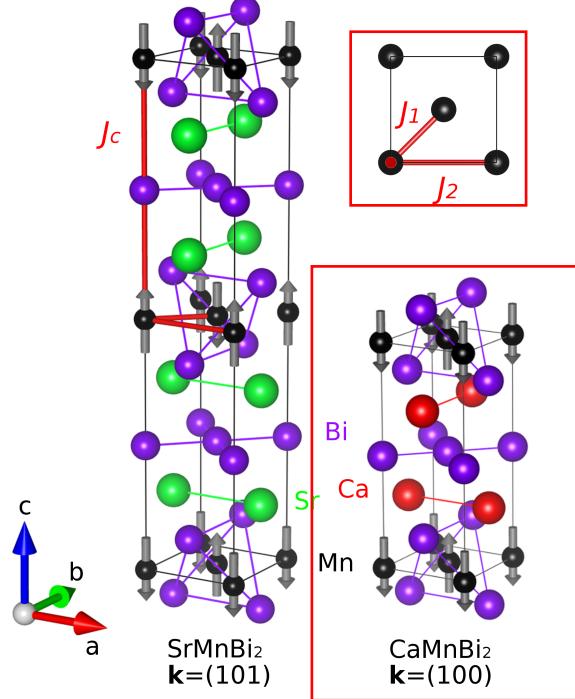


FIG. 1. (color online). The crystal and magnetic structures of SrMnBi_2 ($a \approx 4.58 \text{ \AA}$, $c \approx 23.14 \text{ \AA}$, magnetic space group $I4'/m'm'm'$) and CaMnBi_2 ($a \approx 4.50 \text{ \AA}$, $c \approx 11.07 \text{ \AA}$, magnetic space group $P4'/n'm'm'$)¹¹. The magnetic propagation vectors \mathbf{k} indicated in this figure describe the magnetic structures $\mathbf{m}(\mathbf{r}_j)$ (in lattice coordinates \mathbf{r}_j) by the relation $\mathbf{m}(\mathbf{r}_j) = \mathbf{m}(0) \exp(2\pi i \mathbf{k} \cdot \mathbf{r}_j)$, where $\mathbf{m}(0)$ is the magnetic moment at an arbitrary origin located on a Mn site. For clarity, the origin of the SrMnBi_2 unit cell has been shifted by $(\frac{1}{2}, 0, \frac{1}{4})$ relative to the conventional cell. The exchange paths J_1 , J_2 and J_c are indicated by red lines.

crystals were then grown from self-flux in an alumina crucible. Electron-probe microanalysis confirmed near-ideal stoichiometry, with a small ($\approx 2\%$) Bi deficiency in the Sr compound (for details, see Ref. 11). Laboratory x-ray diffraction measurements confirmed the tetragonal crystal structures reported previously,^{1,2} space groups $I4/mmm$ (SrMnBi_2) and $P4/nmm$ (CaMnBi_2), see Fig. 1. Magnetization measurements on the batch of crystals used here were consistent with previous studies (see supplemental material¹⁴).

Neutron inelastic measurements were performed at the Institut Laue–Langevin on the triple-axis neutron spectrometer IN8 (Ref. 15) with the FlatCone detector.¹⁶ By keeping the outgoing energy fixed and recording rocking scans at various incident energies, this setup allows an efficient collection of constant energy-transfer maps covering a wide range of reciprocal space. The FlatCone array of analyzer crystals and helium tube detectors consists of 31 channels spaced by 2.5° , thus covering a 75° range of scattering angle. Throughout the study, the FlatCone was used with its Si (111) analyzer crystals selecting a fixed outgoing wavevector of $k_f = 3 \text{ \AA}^{-1}$

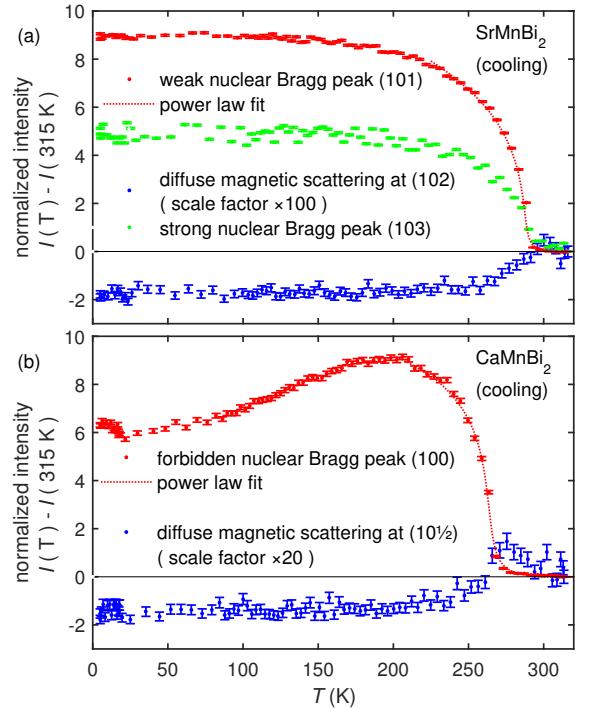


FIG. 2. (color online). Temperature dependence of the difference intensity $I(T) - I(315 \text{ K})$ at selected wavevectors (see legend), recorded while cooling (a) SrMnBi_2 and (b) CaMnBi_2 . Power law fits to the Bragg peaks yield transition temperatures of $T_N^{\text{Sr}} = 287(5) \text{ K}$ and $T_N^{\text{Ca}} = 264(2) \text{ K}$. Above T_N , incipient in-plane correlations contribute diffuse rods of magnetic scattering along $(10L)$. These fluctuations are enhanced towards T_N (critical scattering) and then freeze out with the onset of inter-plane order (blue symbols). The decrease in intensity of the (100) reflection of CaMnBi_2 below 200 K is not consistent with previous data and should be disregarded (see main text).

($E_f = 18.6 \text{ meV}$). For energy transfers below and above 40 meV (incoming energies $E_i \gtrless 58.6 \text{ meV}$), the double-focusing Si (111) and pyrolytic graphite (002) monochromators were used, respectively. In four separate experiments, the scattering from the SrMnBi_2 and CaMnBi_2 single crystals (of mass 3.3 g and 1.6 g, respectively) was investigated in the $(HK0)$ (ab orientation) and $(H0L)$ (ac orientation) scattering planes. Throughout this paper we give wavevectors in reciprocal lattice units (r.l.u.) $\mathbf{q} = (H, K, L) \equiv (H \times 2\pi/a, K \times 2\pi/b, L \times 2\pi/c)$. The samples were mounted in a standard top-loading liquid helium cryostat. All spectra were recorded at a sample temperature of approximately 5 K.

III. RESULTS AND ANALYSIS

While cooling the samples in the ac orientation, we tracked the intensities at selected positions in the $(H0L)$ plane of reciprocal space. Figures 2(a) and (b) show the resulting temperature dependences for SrMnBi_2 and

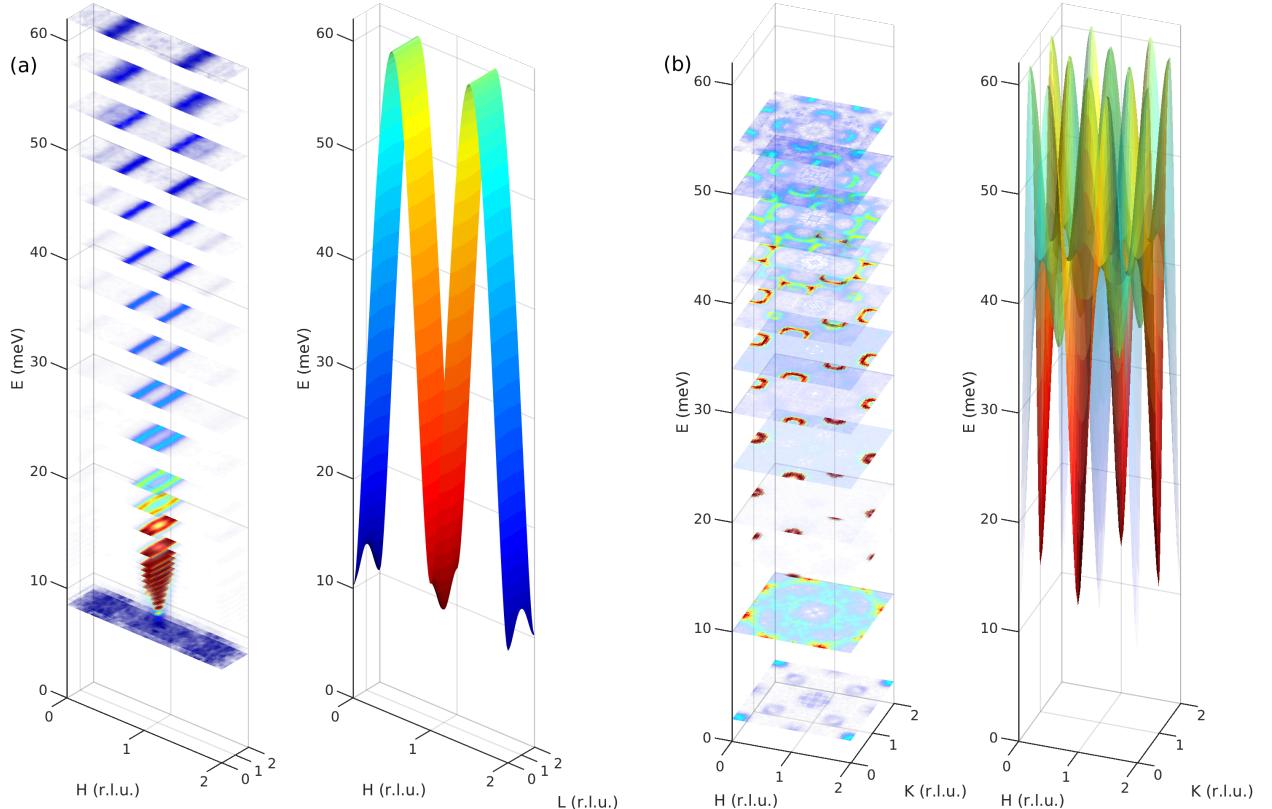


FIG. 3. (color online). Magnon spectrum of SrMnBi_2 in the (a) ($H0L$) and (b) ($HK0$) planes in reciprocal space. The data are illustrated by a stacking plot of constant-energy slices (left panels), and the best-fit spin-wave model is represented by the corresponding dispersion surface (right panels). A more quantitative comparison between data and simulation is provided in the Supplemental Material.¹⁴

CaMnBi_2 , respectively. This includes the magnetic Bragg contribution at (101) and (103) (for Sr) and (100) (for Ca), as well as the diffuse magnetic scattering at another position along the $(10L)$ direction away from the Bragg condition. The data, here represented as the relative change in the square root of the intensity, demonstrate the order parameter characteristics of magnetic Bragg scattering at the antiferromagnetic transitions. We note that the decrease of the CaMnBi_2 (100) magnetic scattering below 200K is not consistent with our previous powder neutron diffraction data,¹¹ and could be due to a shift of the peak between two detector channels as the lattice contracts.

Above the ordering temperature, incipient in-plane magnetic correlations form diffuse rods of magnetic scattering along the c^* direction of reciprocal space, as revealed at the $(10L)$ non-Bragg positions. When cooling towards T_N , this diffuse scattering initially intensifies and then subsides when the weaker inter-layer correlations set in and neutron spectral weight is confined to the Bragg peaks. Fitting a power law to the thermal variation of the (101) (Sr) and (100) (Ca) peaks yields Néel temperatures of $T_N^{\text{Sr}} = 287(5)$ K and $T_N^{\text{Ca}} = 264(2)$ K. These values are consistent with previous single crystal bulk measurements of transport and ARPES samples^{4,9,12,13}, but

differ slightly from the values found in our earlier neutron powder diffraction study¹¹. This difference is likely due to small structural or compositional variations among the samples. The critical exponents $\beta^{\text{Sr}} = 0.15(3)$ and $\beta^{\text{Ca}} = 0.11(2)$ obtained from the power law fit are much smaller than the value $\beta = 0.365$ of the three-dimensional Heisenberg model, indicating the reduced dimensionality of the magnetism in these systems. Due to the additional bismuth layers in the unit cells, the magnetism is more two-dimensional in $AM\text{nBi}_2$ than in the related (122) manganese arsenide BaMn_2As_2 , $\beta = 0.35(2)$ (Ref. 17). Instead, the inter-layer correlations compare well to the parent compounds of (122) iron-based superconductors, e.g. $\beta = 0.098(1)$ for SrFe_2As_2 (Ref. 18) and $\beta = 0.125$ for BaFe_2As_2 (Ref. 19). A detailed description of the power law fit to this data is provided in the Supplemental Material.¹⁴

The measured neutron spectra are summarized in Fig. 3 (SrMnBi_2) and Fig. 4 (CaMnBi_2). A more quantitative presentation of the data is provided in figures S2–S5 of the Supplemental Material.¹⁴ Due to the periodicity of the antiferromagnetic order, the magnetic zone centers are located at (HKL) positions with $(H+K)$ and L both odd integers for SrMnBi_2 , and at positions with $(H+K)$ odd and L any integer for CaMnBi_2 . For both com-

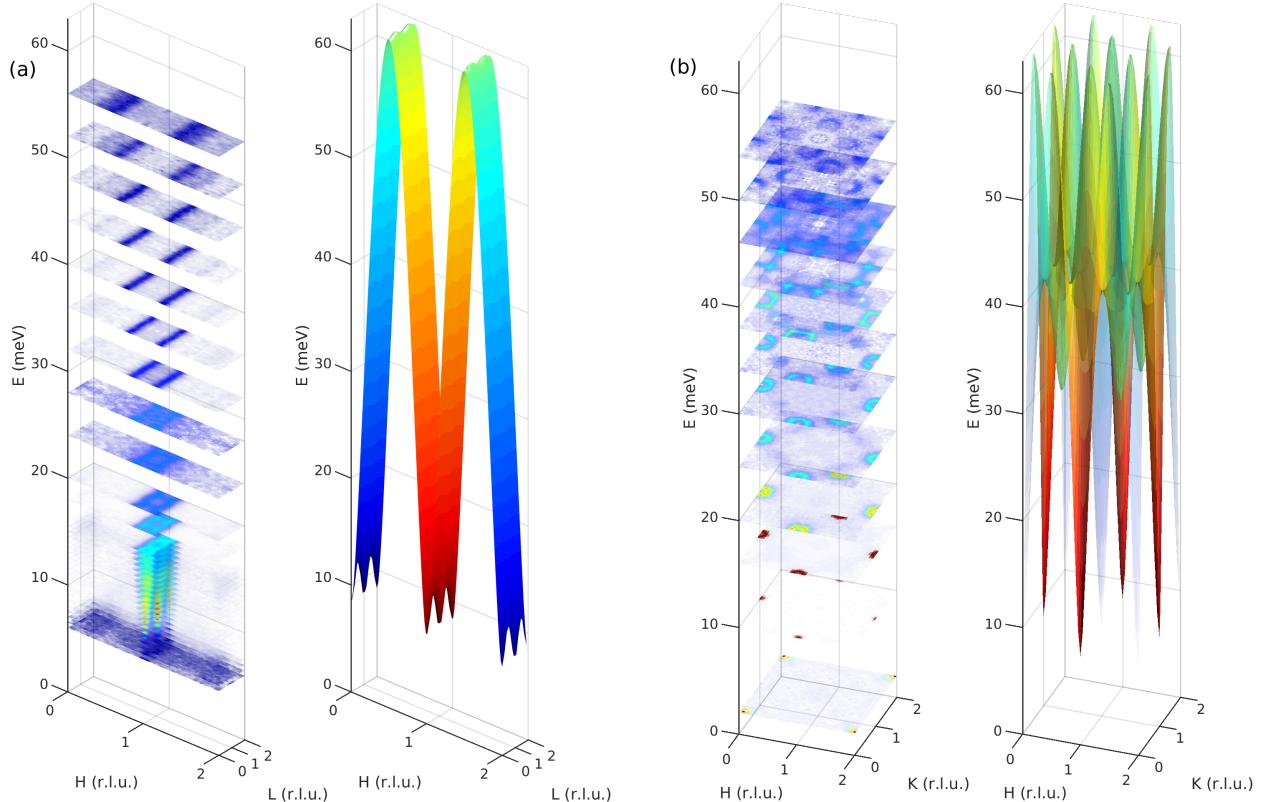


FIG. 4. (color online). Magnon spectrum of CaMnBi_2 presented in the same way as in Fig. 3.

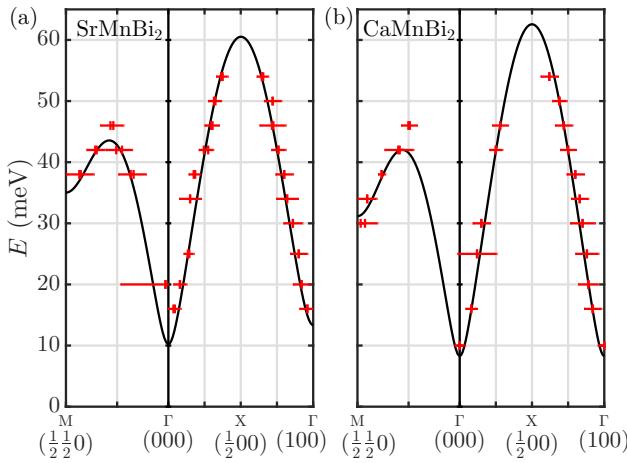


FIG. 5. (color online). Magnon dispersion along high symmetry directions in the $(\bar{H}\bar{K}0)$ plane, for (a) SrMnBi_2 and (b) CaMnBi_2 . The black line indicates the best fit from the linear spin-wave model. Red markers represent the position (vertical bars) and full-width at half-maximum (horizontal lines) of gaussian fits to cuts through the raw data along the corresponding directions.

pounds, the spectra reveal a well-defined magnon dispersion above spin gaps of approximately 10 meV (Sr) and 8 meV (Ca). The magnons are highly dispersive parallel to the layers, but only weakly dispersive perpendicular to

the layers. For both samples the magnon bandwidth is around 50 meV for spin waves propagating in the $(\bar{H}\bar{K}0)$ plane and 3–4 meV along $(10L)$. Figure 5 represents more quantitatively the magnon dispersion in the $(\bar{H}\bar{K}0)$ plane as obtained from gaussian fits to constant-energy cuts, and the left-hand panels of Figs. 6(a) and (b) illustrate the out-of-plane dispersion by energy-wavevector slices of the data along the $(10L)$ direction.

To obtain quantitative information on the magnetic couplings we have compared the data with the linear spin-wave spectrum calculated from an effective spin Hamiltonian that includes a Heisenberg coupling term and an Ising-like single-ion anisotropy,

$$\hat{\mathcal{H}} = \sum_{\langle i,j \rangle} J_{ij} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j - \sum_i D (\hat{S}_i^z)^2, \quad (1)$$

where we include nearest-neighbor (J_1) and next-nearest-neighbour (J_2) exchange constants, an inter-layer exchange interaction J_c , and the anisotropy constant D . The exchange paths are shown in Fig. 1. Using the Holstein–Primakoff transformation of two interacting Bose fields, corresponding to the two collinear antiferromagnetic sublattices, we obtain the dispersion relation

$$E(\mathbf{q}) = \sqrt{A(\mathbf{q})^2 - B(\mathbf{q})^2} \quad (2)$$

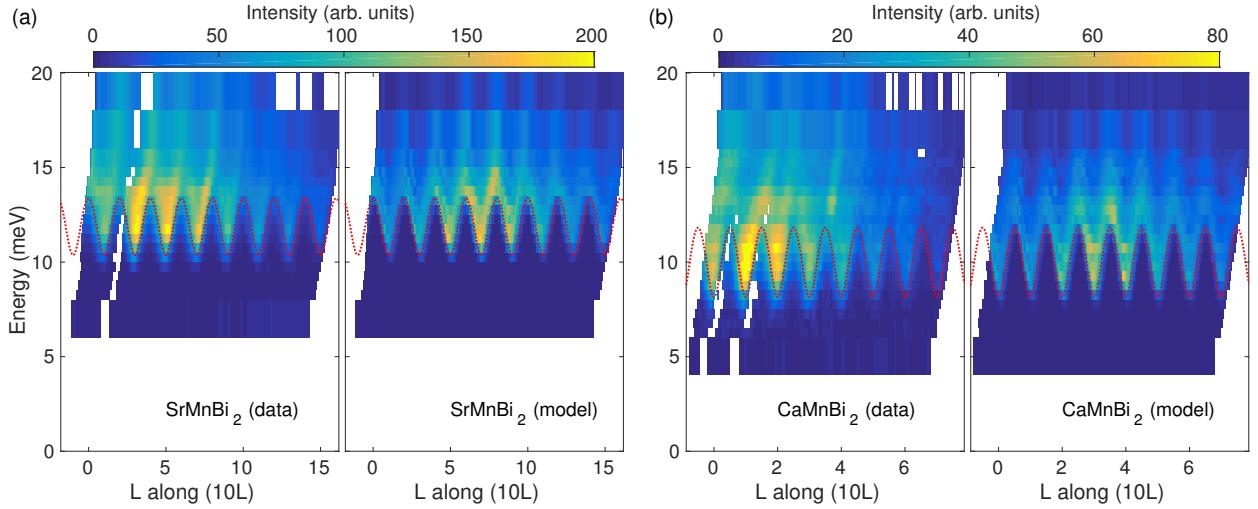


FIG. 6. (color online). Out-of-plane dispersion of the magnon spectra of (a) SrMnBi₂ and (b) CaMnBi₂, illustrated by slices along the (10L) direction in reciprocal space (intensity averaged over the range $H = 0.95\text{--}1.05$ r.l.u.). The left-hand panels show interpolated plots of the data, and the right-hand panels give the corresponding best-fit spin-wave spectra convoluted with the instrumental resolution (for details, see text and Supplemental Material¹⁴). The superimposed red dashed line indicates the theoretical dispersion using best-fit parameters.

where \mathbf{q} is the magnon wavevector,

$$\begin{aligned} A(\mathbf{q}) &= S [\mathcal{J}_{\text{AF}}(0) - \mathcal{J}_{\text{F}}(0) + \mathcal{J}_{\text{F}}(\mathbf{q}) + 2D] \\ B(\mathbf{q}) &= S \mathcal{J}_{\text{AF}}(\mathbf{q}) \end{aligned}$$

and

$$\mathcal{J}(\mathbf{q}) = \sum_n J_n e^{2\pi i \mathbf{q} \cdot \mathbf{r}_n} \quad (3)$$

is the Fourier transform of the exchange interactions. The subscripts F and AF refer to summation over ferromagnetically- and antiferromagnetically-aligned spins, respectively. The resulting differential scattering cross-section for single-magnon creation is

$$\frac{d\sigma}{d\Omega d\omega} = \frac{k_f}{k_i} \left(\frac{\gamma r_0}{2} \right)^2 S(\mathbf{q}, \omega) \quad (4)$$

$$S(\mathbf{q}, \omega) = g^2 N S \frac{A(\mathbf{q}) - B(\mathbf{q})}{E(\mathbf{q})} \{n(\omega) + 1\} \delta\{\hbar\omega - E(\mathbf{q})\} \quad (5)$$

where $\hbar\omega$ is the neutron energy transfer, k_f and k_i are the outgoing and incoming neutron wave vectors, $\gamma = 1.913$, r_0 is the classical electron radius, g the Landé g -factor, N the number of magnetic ions per sublattice, S the spin quantum number, and $n(\omega) = (e^{\hbar\omega/k_B T} - 1)^{-1}$ the boson occupation number. Given the magnetic structures and exchange paths defined in Fig. 1, the explicit Fourier exchange terms for the case of SrMnBi₂ are

$$\begin{aligned} \mathcal{J}_{\text{AF}}^{\text{Sr}}(\mathbf{q}) &= 2 J_1 [\cos(\pi H + \pi K) + \cos(\pi H - \pi K)] + \\ &\quad + 2 J_c \cos(\pi L) \end{aligned}$$

$$\mathcal{J}_{\text{F}}^{\text{Sr}}(\mathbf{q}) = 2 J_2 [\cos(2\pi H) + \cos(2\pi K)]$$

and, in the case of CaMnBi₂,

$$\begin{aligned} \mathcal{J}_{\text{AF}}^{\text{Ca}}(\mathbf{q}) &= 2 J_1 [\cos(\pi H + \pi K) + \cos(\pi H - \pi K)] \\ \mathcal{J}_{\text{F}}^{\text{Ca}}(\mathbf{q}) &= 2 J_2 [\cos(2\pi H) + \cos(2\pi K)] + 2 J_c \cos(2\pi L). \end{aligned}$$

This allows an analytical description of the spin gaps:

$$\Delta^{\text{Sr}} \approx \Delta^{\text{Ca}} \approx 4 \sqrt{SJ_1} \sqrt{SD}, \quad (6)$$

where we have applied the appropriate approximations for the present case ($J_1 \gg J_c$, $J_1 \gg D$; for full expressions see the Supplemental Material¹⁴). Similarly, the band width W of the dispersion along (10L) is given by

$$W^{\text{Sr}} \approx W^{\text{Ca}} \approx 4 \sqrt{SJ_1} \left(\sqrt{SD + 2|SJ_c|} - \sqrt{SD} \right). \quad (7)$$

If J_1 is the dominant exchange, as is found to be the case here, then the maximum of the in-plane dispersion is $\sim 4SJ_1$. Given J_1 , we see from Eqs. (6) and (7) that in the relevant parameter regime the parameters D and J_c are determined by the size of the gap $\Delta^{\text{Sr/Ca}}$ and band width of the out-of-plane modulation $W^{\text{Sr/Ca}}$, respectively. On the other hand, the balance between the parameters J_1 and J_2 determines details of the dispersion at higher energies in the (HK0) plane. For example, a local minimum of the dispersion at the M point, $(\frac{1}{2}, \frac{1}{2}, 0)$, as observed in both materials, will only occur for positive J_2 , indicating a competition (frustration) between nearest- and next-nearest-neighbor exchange.

We find that the above model is able to reproduce very well all features in the data. For quantitative analysis we folded and averaged the raw constant-energy maps of reciprocal space into tiles of 2×2 r.l.u. With the data in this reduced form we could compare it to the model after convolution of the theoretical spectrum, Eqs. (4)–(5),

with an energy- and wavevector-dependent broadening function to take into account the instrumental resolution.

A phenomenological gaussian broadening of the analytical dispersion proved insufficient to achieve a consistent global fit to the data, particularly for the low-energy part of the magnetic dispersion in the *ac* plane. Instead, it was necessary to take into account the resolution of the triple-axis spectrometer, which was calculated with the RESTRAX ray-tracing algorithm^{20,21}. Our procedure to determine the parameters of the spin Hamiltonian J_1 , J_2 , J_c and D was carried out in three steps: First, a global fit of all data, using phenomenological gaussian broadening of the dispersion, produced rough estimates of all parameters. Using these as starting values and fixing the in-plane exchange interactions J_1 and J_2 , we obtained precise bounds on the inter-layer exchange J_c and anisotropy D by fitting the resolution-convoluted spectrum for low energies (0–20 meV) to an energy–wavevector slice with wavevector along $(10L)$, as illustrated in Fig. 6. Finally, J_1 and J_2 were refined by fitting the in-plane (*ab*) dispersion at high energies (30–44 meV) using gaussian broadening.

Figure 5 provides a quantitative plot of the fits of the dispersions in the *ab*-plane. A more detailed description of the data processing, fitting and error estimation, and an explicit comparison of the data and best fits is provided in the Supplemental Material.¹⁴

IV. DISCUSSION

The exchange parameters for SrMnBi₂ and CaMnBi₂ obtained from the fits are summarized in Table I. Apart from the opposite sign of the inter-layer exchange J_c , there are no significant differences between the parameters of SrMnBi₂ and CaMnBi₂. The absolute values of J_1 and J_2 are slightly larger in the case of CaMnBi₂, consistent with the smaller nearest-neighbor spacing (d_{NN}). The magnitudes of J_c for the two compounds, which are the same to within the error, are much smaller than J_1 and J_2 , confirming the quasi-2D character of the magnetism in these materials. Notably, these results are in good agreement with previous estimations based on first principles calculations of the electronic structure, which gave an average in-plane exchange of $SJ_{ab} \approx 30$ meV and $|SJ_c| \approx 0.3$ meV¹¹.

Regarding the magnetocrystalline anisotropy, we observe that D is enhanced by a factor 1.8 in SrMnBi₂ compared with CaMnBi₂. According to the initial structure determinations at room temperature^{1,2}, the local environment of the Mn ion is similar in both compounds: The MnBi₄ tetrahedra are elongated by $\approx 14\%$ along c and the ligand distances are $d_{\text{Mn-Bi}}^{\text{Ca}} = 2.87(1)$ Å and $d_{\text{Mn-Bi}}^{\text{Sr}} = 2.89(1)$ Å. The significant difference in anisotropy may therefore point to unknown structural distortions at 5 K (at present, no full refinement of crystallographic parameters at low temperatures is available). The anisotropy is in good agreement with the result of our earlier density

functional prediction ($SD_{\text{DFT}}^{\text{Ca}} = 0.3$ meV¹¹), as was also the case with the exchange constants.

It is instructive to compare the present results to two available inelastic neutron studies of the related compounds BaMn₂Bi₂²² and BaMn₂As₂²³. The corresponding parameters for these materials are quoted in Table I. The pnictide-coordinated magnetic Mn²⁺ layers in BaMn₂Bi₂ and BaMn₂As₂ (“122 materials”) are analogous to those in the 112 materials investigated in the present study. However, the $I4/mmm$ 122 compounds do not feature additional pnictide layers (which carry the Dirac bands in the present case). Hence, while the in-plane Mn–Mn spacing is very similar, the spacing of the magnetic layers in the 122 compounds is only 58–66% of that in CaMnBi₂ and SrMnBi₂. Both BaMn₂Bi₂ and BaMn₂As₂ form antiferromagnetically stacked layers of Néel type order, in analogy to SrMnBi₂. As may be expected from these circumstances, we find that the in-plane exchange interactions in 122 compounds are similar or identical to those in 112 compounds. On the other hand, in the present 112 materials the inter-plane exchange is significantly reduced. This is consistent with the much higher transition temperatures and the smaller separation of the Mn layers in the 122 materials compared with the 112 compounds.

We find no evidence that the additional Bi layers in 112 materials, which host the Dirac fermions, cause any qualitative changes in the magnon spectrum, such as anomalous broadening or dispersion. The instrument’s simulated energy resolution provides an upper bound on the influence of such effects. The characteristics of the Bragg (0.5–1.0 meV) and vanadium (1–4 meV) widths of energy resolution are illustrated in the Supplemental Material.¹⁴ By contrast, neutron inelastic measurements of many iron-based superconductors show obvious signatures of a strong hybridization of magnetic and itinerant states. A typical example is SrFe₂As₂²⁵, which shows a crossover into the regime of itinerant (*Stoner*) spin fluctuations. This manifests itself as an increased dampening of spin fluctuations (i.e. a broadening of the neutron spectrum) above a characteristic energy of approximately 80 meV.

As in the 122 compounds, both J_1 and J_2 are positive (antiferromagnetic) in SrMnBi₂ and CaMnBi₂, resulting in frustration between nearest- and next-nearest-neighbor interactions. The theoretical phase diagram of the frustrated J_1 – J_2 model on a square lattice has been investigated extensively in the context of iron-based superconductors.^{26–28} There is special interest in this phase diagram owing to a possible quantum critical point and spin liquid phase around $J_2/J_1 \approx \frac{1}{2}$. This regime separates two distinct ordered magnetic phases, with Néel type order for $J_2/J_1 < \frac{1}{2}$ and stripe antiferromagnetic order for $J_2/J_1 > \frac{1}{2}$. Both 112 and 122 Mn-based compounds exhibit dominant nearest-neighbor exchange, with $J_2/J_1 \approx 0.3$. According to one study the exchange and anisotropy parameters for AMnBi₂ places these materials close to the phase boundary between Néel-ordered and frustrated paramagnetic phases.²⁸ The resulting

TABLE I. Exchange parameters, magnetocrystalline anisotropy constants and spin gaps of SrMnBi₂ and CaMnBi₂ obtained from a fit of the linear spin-wave model, as described in the text. The parameters can be related to the nearest-neighbor (d_{NN}) and interlayer (d_c) Mn–Mn atomic spacings, the ordered magnetic moment μ , and the ordering temperature T_N .¹¹ The corresponding values for two related Mn pnictides are reproduced below.^{22,23}

	SJ_1 (meV)	SJ_2 (meV)	SJ_c (meV)	SD (meV)	Δ (meV)	d_{NN} (Å)	d_c (Å)	$\mu(\mu_B)$	T_N (K)
SrMnBi ₂	21.3(2)	6.39(15)	0.11(2)	0.31(2)	10.2(2)	3.24	11.57	3.75(5)	290.2(3)
CaMnBi ₂	23.4(6)	7.9(5)	-0.10(5)	0.18(3)	8.3(8)	3.18	11.07	3.73(5)	267.0(1.6)
BaMn ₂ Bi ₂ [22]	21.7(1.5)	7.85(1.4)	1.26(2)	0.87(15) ^a	16.29(26)	3.18	7.34	3.83(4)	387.2(4)
BaMn ₂ As ₂ [23]	33(3)	9.5(1.3)	3.0(6)	-	-	2.95	6.73	3.88(4)	625(1)

^a The value of SD for BaMn₂Bi₂ was misquoted in Ref. 22. In this table we give the correct value²⁴.

quantum fluctuations could explain some of the observed reduction in ordered magnetic moment ($\simeq 3.7 \mu_B$) compared to the ideal local-moment value of $5 \mu_B$ ²³. By contrast, in parent compounds of iron-based superconductors such as BaFe₂As₂ and SrFe₂As₂, J_1 and J_2 are of similar magnitude, resulting in stripe-antiferromagnetic order.

V. CONCLUSIONS

In summary, we have performed a comprehensive triple-axis neutron scattering study of the anisotropic Dirac materials SrMnBi₂ and CaMnBi₂, with the aim of searching for possible influences of the unusual band topology at their Fermi surfaces on their magnetism. In particular, for CaMnBi₂ our previous findings had indicated that the Bi $6p_{x,y}$ bands may play a role in mediating the magnetic exchange between Mn layers.

In both compounds, we observed well-defined magnon spectra consistent with local-moment, semi-classical antiferromagnetism. Using linear spin-wave theory to describe the neutron spectra we have identified and quantified all relevant exchange and anisotropy parameters of a Heisenberg model for the two compounds. In both cases, all details of the dispersion are well reproduced by the model and there is no indication of anomalous broadening or dispersion to within experimental precision. The absolute values of the exchange parameters indicate no substantive differences between the compounds (aside from opposite interlayer coupling).

These results suggest that different routes have to be found to achieve an entanglement of magnetic order and non-trivial band topology. One very promising option is the substitution of magnetic rare earth ions on the A site, providing a more direct interaction with the relevant Bi layers. In particular, a strong response of the transport properties to rare earth magnetic order has recently been observed in EuMnBi₂²⁹, along with the trademark signatures of Dirac transport³⁰. Furthermore, recent high-resolution ARPES results and first principles calculations identify YbMnBi₂ as a type-2 Weyl semimetal with canted antiferromagnetic order³¹. The latter study further suggests that this state would be tuned to a Dirac

metal by spin alignment. Naturally, it would be of great interest to perform analogous inelastic neutron studies of the magnetic ground states in those materials.

Note added in proof: After submission of this manuscript, a Raman spectroscopic study of SrMnBi₂ and CaMnBi₂ was reported by Zhang *et al.*³². Raman spectroscopy probes the spin dynamics through a small number of characteristic frequencies which are associated with van-Hove singularities in the two-magnon density of states. The authors of Ref. 32 interpret their data using a similar spin Hamiltonian as in the present study but without the magnetocrystalline anisotropy term (D in our study). Their analysis yields values for the spin exchange parameters J_1 and J_2 that are similar to our results, but produces anomalously large values of the interlayer exchange J_c for both materials (one order of magnitude larger than in our study or in other related materials). The authors of Ref. 32 suggest that this enhanced coupling is caused by the Bi Dirac bands. We would like to draw attention to the fact that the parameters J_1 , J_c and D are strongly correlated in modelling key features of the magnon dispersion (see Eqs. 6 and 7), so the neglect of D in the Raman analysis could significantly affect the obtained values of J_c . We note that the Raman value of J_c would imply an inter-layer dispersion of the one-magnon spectrum at a factor of 11 (Sr) or 7 (Ca) larger than that found here directly by neutron spectroscopy (Fig. 6, Eq. 7).

ACKNOWLEDGMENTS

We would like to thank Dr. Paul Steffens (ILL) for providing the FLATCONE data treatment suite, and Dr Stuart Calder for helpful clarifications on the results reported in Ref. 22. This work was supported by the U.K. Engineering and Physical Sciences Research Council (grant no. EP/J017124/1), the Chinese National Key Research and Development Program (2016YFA0300604) and the Strategic Priority Research Program (B) of the Chinese Academy of Sciences (Grant No. XDB07020100). MCR is grateful to the Oxford University Clarendon Fund for provision of a scholarship.

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Supplemental Material:

Spin dynamics in the antiferromagnetic phases of the Dirac metals $AMnBi_2$ ($A = Sr, Ca$)

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1. Power law fit of magnetic Bragg peaks

As described in the manuscript, the thermal variation of the intensities of the (101) ($SrMnBi_2$) and (100) ($CaMnBi_2$) magnetic peaks were fitted with a power law. To reproduce the incipient fluctuations above T_N , the power law fitting function was convoluted by a Lorentzian distribution of the ordering temperature:

$$I \propto A(T_N - T)^{2\beta} * \frac{1}{\gamma^2 + (T - T_N)^2}.$$

The five (unconstrained) fitting parameters were then (1) a constant background, (2) an overall scale factor A , (3) the critical exponent β , (4) the Néel temperature T_N and (5) the Lorentzian full width at half maximum γ (see Fig. S1). While T_N always converges to the results quoted in the main text, the critical exponents β weakly depend on the selected fitting range. In the main text we therefore quote the mean fit results and standard deviations of separate fits to data ranges varying between $(T_N - 65 K)$ to 313 K and $(T_N - 5 K)$ to 313 K.

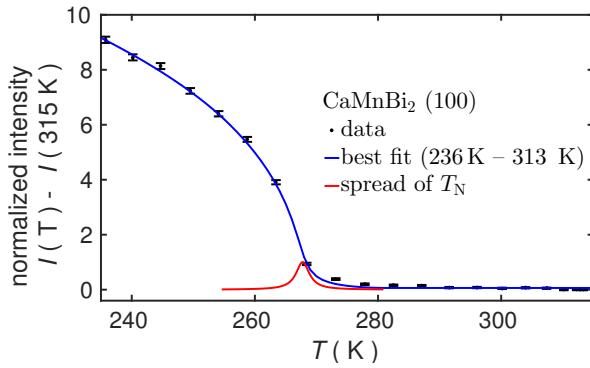


Fig. S1 (color online). Lorentzian-convoluted power law fit to the relative thermal variation of the scattering amplitude at the magnetic Bragg reflection (100) of $CaMnBi_2$.

2. Magnetic susceptibility

To check for consistency with our previous study¹¹, we measured the thermal variation of the magnetic properties of the batch of samples probed by neutron inelastic scattering (see Fig. S2). Single crystals were first aligned on an x-ray diffractometer. The magnetic susceptibility

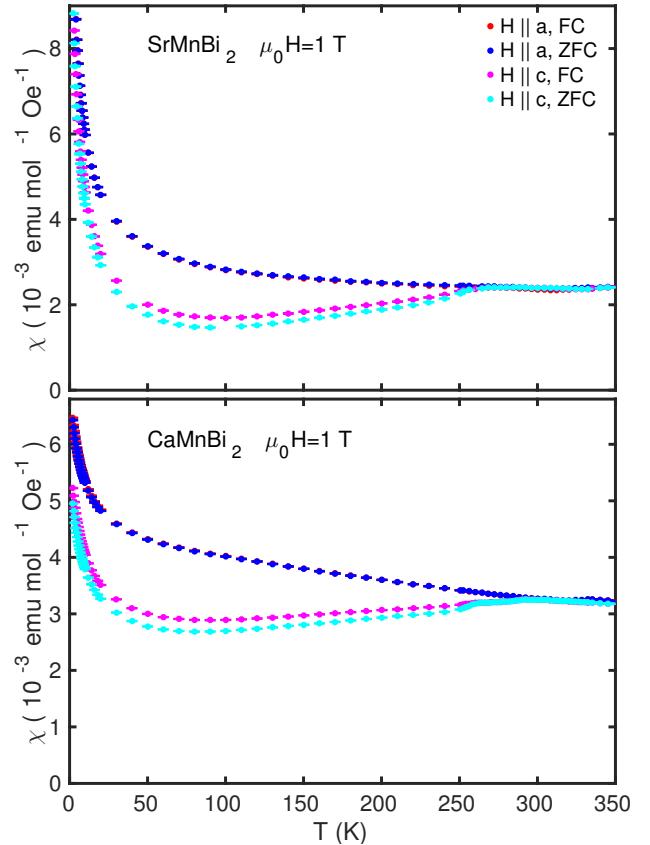


Fig. S2 (color online). Magnetic susceptibility of $SrMnBi_2$ (top) and $CaMnBi_2$ (bottom), measured on the batch of single crystals probed by neutron spectroscopy.

was then measured on a Magnetic Properties Measurement System (Quantum Design) with a magnetic field of flux density 1 Tesla applied either in-plane or out-of-plane. The key characteristics are qualitatively consistent with our earlier results¹¹. The present samples have a larger Curie contribution which may be attributed to paramagnetic impurities induced by sample decay. Since the magnon dispersion is a coherent response of the main crystal phase, such impurities are not of relevance to the present study, apart from a small contribution to the diffuse background scattering.

3. Analytical expressions for features in the magnon dispersion

The linear spin-wave model yields the following analytical expressions for the spin gaps:

$$\Delta^{\text{Sr}} = S [(4J_1 + 2J_c + 2D)^2 - (4J_1 + 2J_c)^2]^{\frac{1}{2}} \approx 4\sqrt{SJ_1} \sqrt{SD}$$

$$\Delta^{\text{Ca}} = S [(4J_1 + 2D)^2 - (4J_1)^2]^{\frac{1}{2}} \approx 4\sqrt{SJ_1} \sqrt{SD},$$

where we have applied the appropriate approximations for the present case ($J_1 \gg J_c$; $J_1 \gg D$). Similarly, for the bandwidth W of the dispersion along $(10L)$ we obtain

$$W^{\text{Sr}} = S [(4J_1 + 2J_c + 2D)^2 - (4J_1 - 2J_c)^2]^{\frac{1}{2}} - S [(4J_1 + 2J_c + 2D)^2 - (4J_1 + 2J_c)^2]^{\frac{1}{2}} \approx 4\sqrt{SJ_1} (\sqrt{SD + 2SJ_c} - \sqrt{SD})$$

$$W^{\text{Ca}} = S [(4J_1 - 4J_c + 2D)^2 - (4J_1)^2]^{\frac{1}{2}} - S [(4J_1 + 2D)^2 - (4J_1)^2]^{\frac{1}{2}} \approx 4\sqrt{SJ_1} (\sqrt{SD - 2SJ_c} - \sqrt{SD}).$$

4. Analysis of neutron spectra

For both materials, and for each in two crystal orientations (momentum transfers in the $(HK0)$ and $(H0L)$ planes of reciprocal space), rocking scans were recorded at a number of energy transfers up to 60 meV. The scattering was recorded in the FlatCone analyser-detector assembly¹⁶ producing a set of constant-energy maps. In each rocking scan the wide FlatCone detector bank is mapped onto an arc in reciprocal space intercepting several Brillouin zones. The spectral weight in these maps shows contributions from Bragg diffraction (at $E = 0$ meV), acoustic phonons (below $E \approx 10$ meV) and magnons (above $E = 8$ –10 meV). In addition, for

$E \neq 0$ meV, the data contain accidental Bragg reflections (i.e. the strong elastic signal passes through the analyzer by parasitic scattering), as well as powder rings corresponding to Bragg diffraction by the sample environment. The magnon spectral weight can clearly be distinguished from the phonon- and accidental Bragg scattering in every dataset from the form of the scattering and from the reduction in the magnetic signal with $|\mathbf{q}|$.

To allow a direct comparison with the model, each raw dataset was corrected in several steps. First, all intensity other than a diffuse background and aluminium powder rings were masked. This masked data was then radially averaged (along the rocking angle). Next, both the raw dataset and the radially averaged powder/background dataset were divided by the magnetic form factor of Mn^{2+} . After subtraction, the trajectories of intense spurious (parasitic) Bragg scattering were deleted from the data manually. The resulting corrected constant-energy maps were then interpolated to a regular grid and divided into equivalent tiles of 2×2 reciprocal lattice units (r.l.u.). Finally, the arithmetic mean of these cells was calculated and all relevant symmetries (mirror planes and 4- or 2-fold rotation of the $(HK0)$ and $(H0L)$ reciprocal lattice planes, respectively) were applied in order to distil the full statistical significance of the data. The resulting constant-energy intensity maps for both compounds and both sample orientations are shown in Figs. S4–S7.

5. Fitting procedure

The constant-energy maps thus obtained are in a convenient form for fitting to the spin-wave model. However, in particular for the low energy part of the magnetic dispersion, a phenomenological gaussian broadening of the analytical dispersion (along the reciprocal space and energy dimensions) proved insufficient. Instead, it was necessary to take into account the resolution of the triple-axis spectrometer. This was done by numerically simulating four-dimensional resolution matrices using the RESTRAX ray tracing algorithm^{20,21}. This algorithm takes into account a full physical model of all relevant components (including collimators, monochromator, analyzer, detectors) of the IN8 instrument. To calculate the resolution matrix for a particular instrument configuration, the program was set to trace 5000 random neutron events. The resulting cloud of (H, K, L, E) positions was fitted to a four-dimensional ellipsoid.

The energy resolution of the triple-axis spectrometer depends only weakly on the momentum transfer, but increases with energy transfer. While the energy Bragg-width of the resolution ellipsoid remains smaller than 1 meV (standard deviation) up to $\Delta E = 60$ meV, the vanadium width increases up to ≈ 4 meV in this range. Figure S3 illustrates these characteristics, which represent the upper bounds within which we can exclude anomalies in the dispersion.

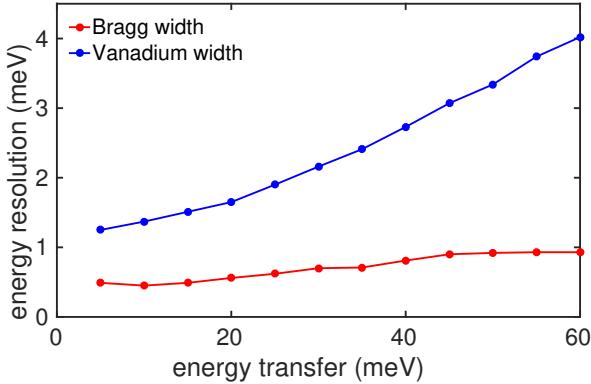


Fig. S3 (color online). Simulated energy resolution of the IN8 spectrometer (RESTRAX algorithm) for the experimental conditions of the present study.

In order to simulate the effect of the resolution on the distribution of magnetic intensity, the resolution ellipsoids were calculated for every pixel of a full dataset. For a particular set of model parameters, the contribution to the intensity at a pixel was calculated by performing a four-dimensional convolution of the ellipsoid with the analytical dispersion. Finally, the so-obtained resolution-corrected intensity corresponding to the full experimental dataset was folded into 2×2 r.l.u. tiles as described for the raw data above.

While this procedure produces a satisfactory simulation of the data, it is unfortunately computationally

too intensive to include in an efficient global fitting routine. The best-fit parameters were therefore determined in three stages: First, a global fit (with phenomenological broadening) of all available datasets (all energies, both orientations) was used to estimate rough starting values for all parameters. Secondly, for an appropriate grid of J_c and D values, resolution-convoluted constant-energy maps corresponding to the low energy ($E \leq 20$ meV) data sets were calculated. Momentum transfer slices along the $(10L)$ direction were then calculated for both data and model. The χ^2 maps resulting from this comparison are shown in Figs. S8(a) and (c) for SrMnBi₂ and CaMnBi₂, respectively, with the contour of one standard deviation indicated by a red line. The best fit $(10L)$ slices thus determined are illustrated in Fig. 6 of the main article.

Finally, with J_c and D fixed, global fits of the high energy dispersion ($30 \leq E \leq 45$ meV, both $HK0$ and $H0L$ orientations) were performed for an appropriate grid of J_1 and J_2 values, using a phenomenological broadening of the dispersion. The resulting χ^2 maps are shown in Fig. S8(b) and (d) for SrMnBi₂ and CaMnBi₂, respectively. The J_1 and J_2 parameters are seen to be strongly correlated in both cases, with a minimum in χ^2 extending along a straight line. However, although χ^2 does not vary significantly along this line, there are weak features in the high energy ($HK0$) maps which do depend sensitively on (J_1, J_2) along this line. These features enabled us to establish empirically the errors on J_1 and J_2 quoted in the main article and indicated by dashed lines in Fig. S8.

Figures S4–S7 give a comparison of all the data with the corresponding simulations from the best-fit model.

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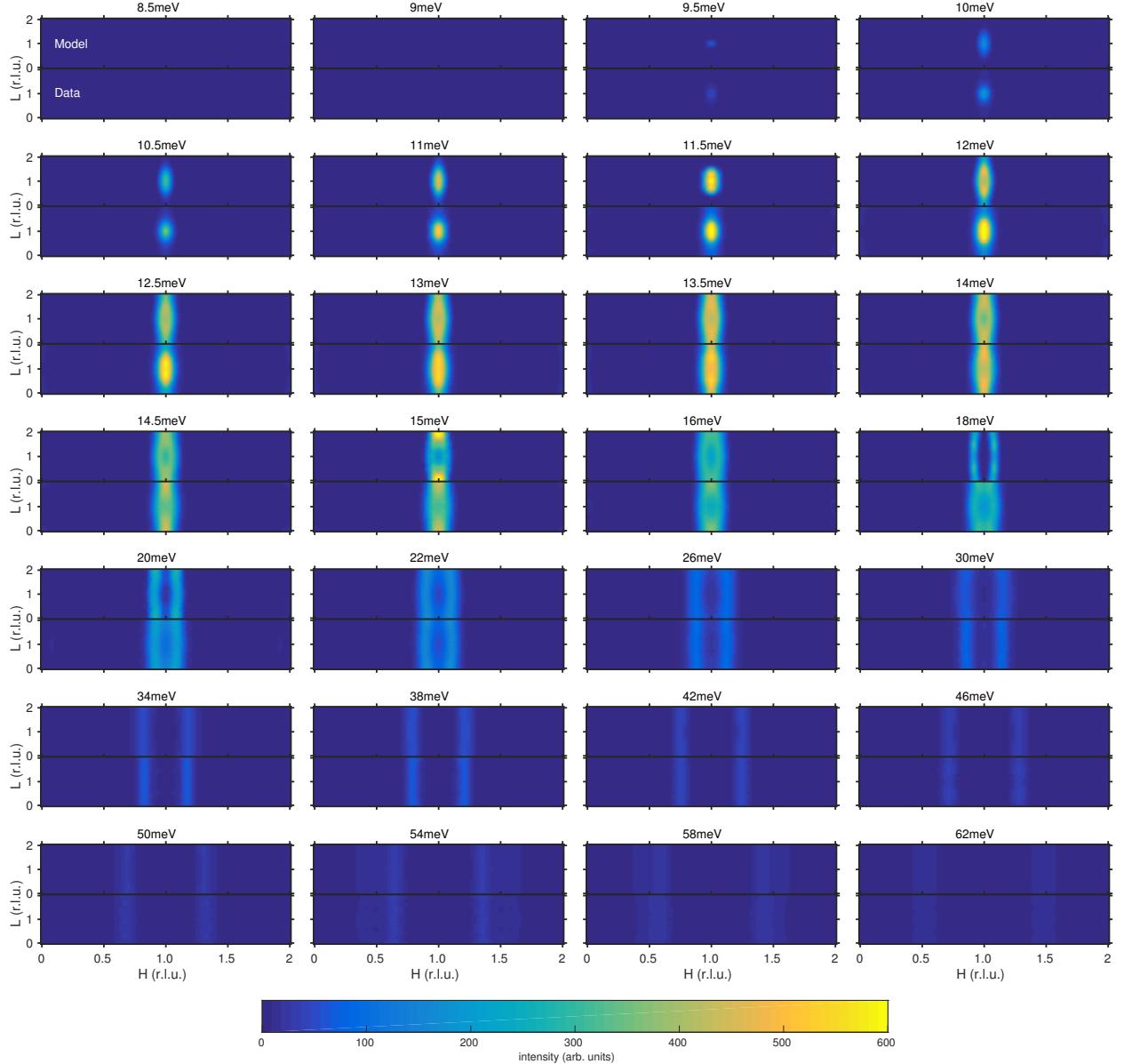


Fig. S4 (color online). SrMnBi₂: Constant-energy maps of a 2×2 r.l.u. area of the ($H0L$) plane of reciprocal space. Each double panel shows the processed data (lower panel, explanation see text) as well as the best fit with a phenomenological gaussian broadening (upper panel).

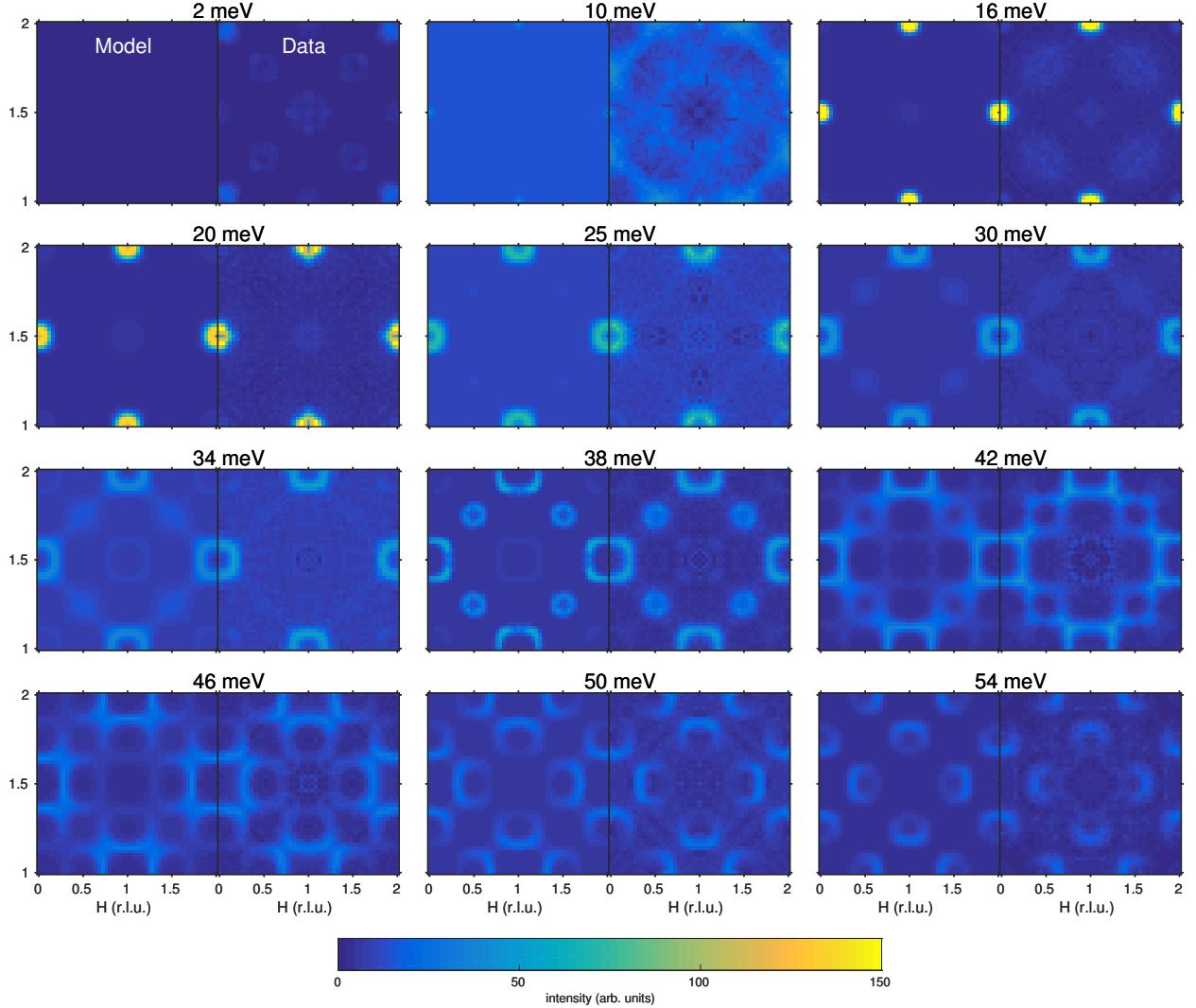


Fig. S5 (color online). SrMnBi₂: Constant-energy maps of a 2×2 r.l.u. area of the $(HK0)$ plane of reciprocal space. Each double panel shows the processed data (right panel, explanation see text) as well as the best fit with a phenomenological gaussian broadening (left panel).

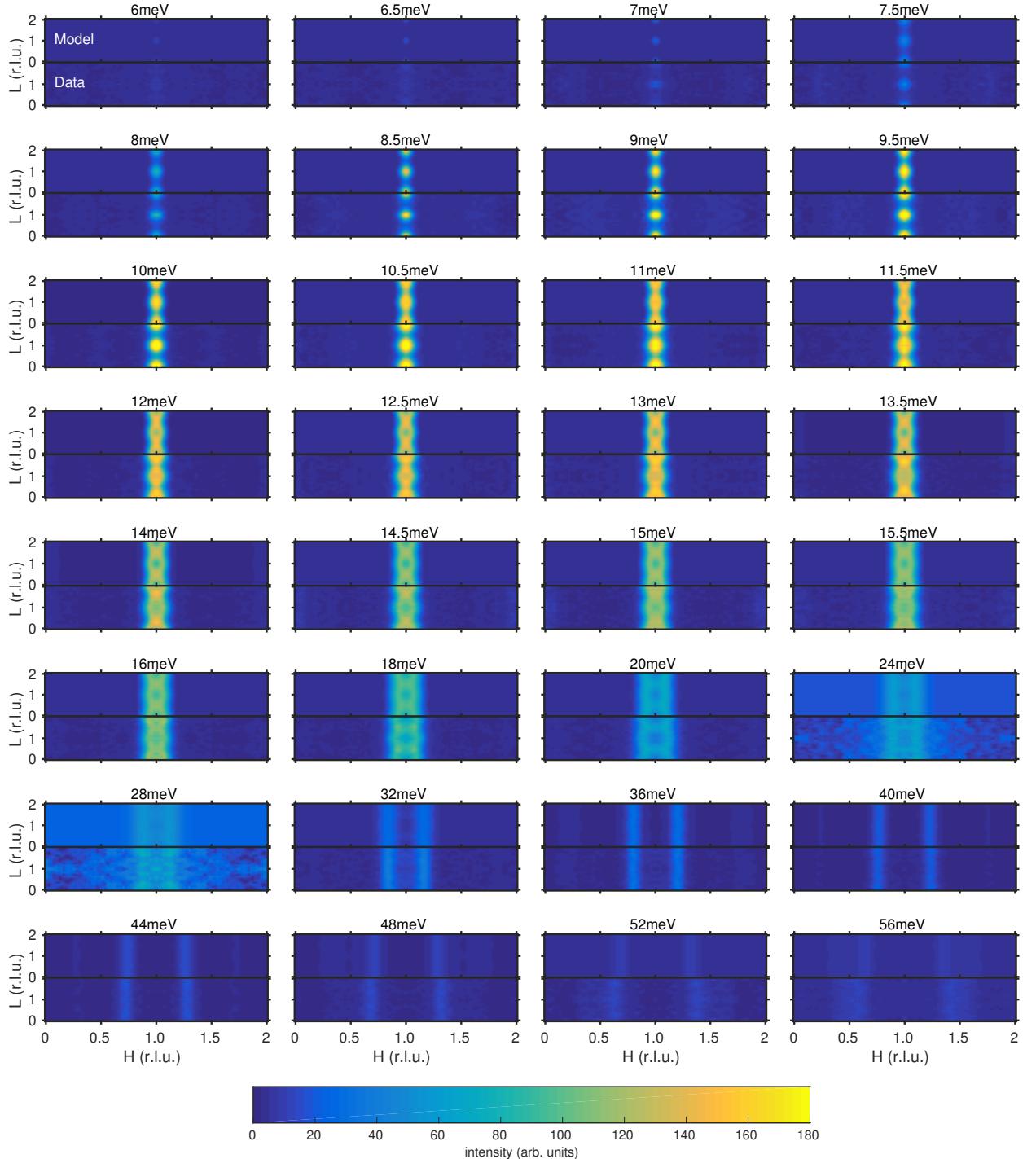


Fig. S6 (color online). CaMnBi₂: Constant-energy maps of a 2×2 r.l.u. area of the ($H0L$) plane of reciprocal space. Each double panel shows the processed data (lower panel, explanation see text) as well as the best fit with a phenomenological gaussian broadening (upper panel).

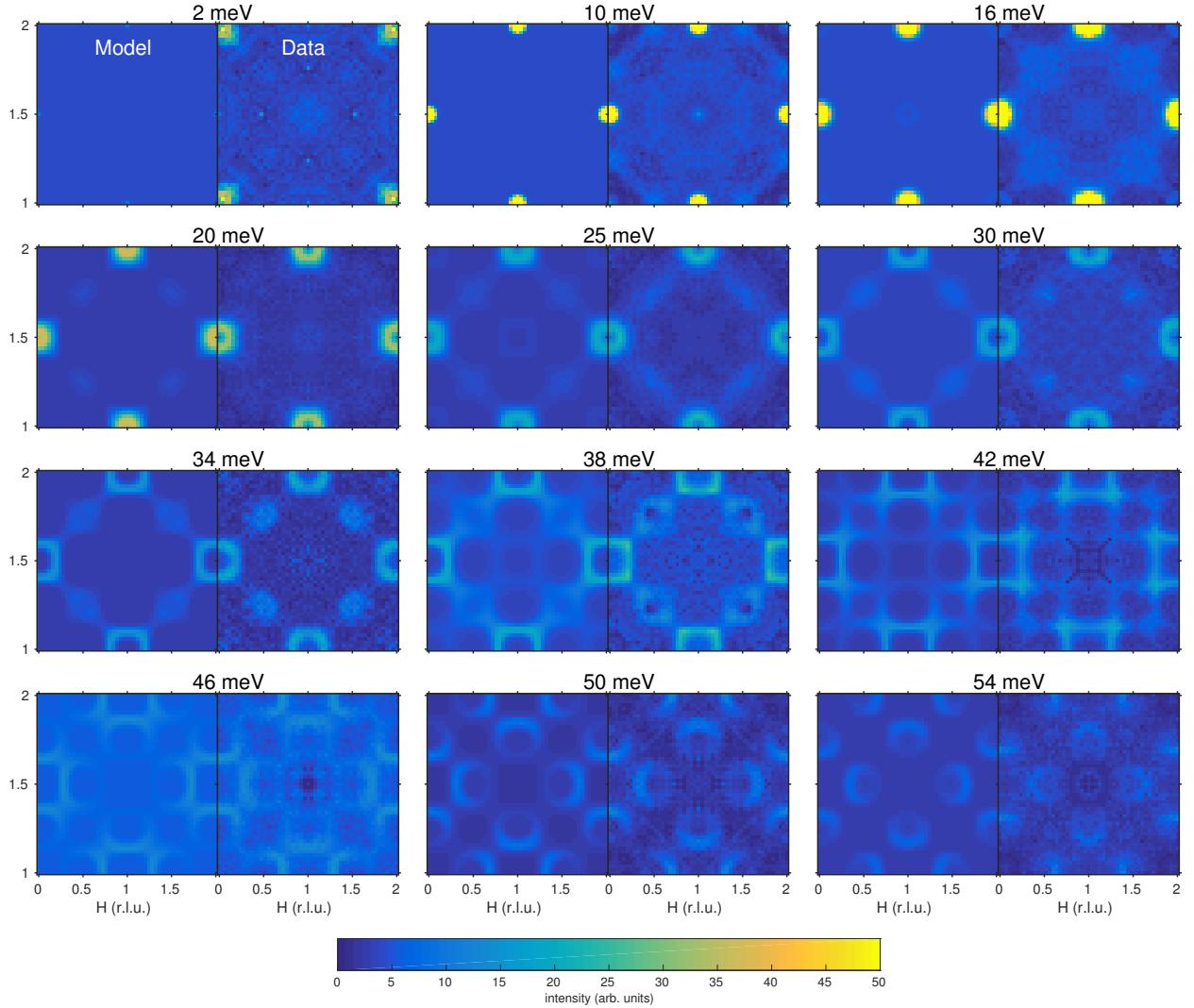


Fig. S7 (color online). CaMnBi₂: Constant-energy maps of a 2×2 r.l.u. area of the $(HK0)$ plane of reciprocal space. Each double panel shows the processed data (right panel, explanation see text) as well as the best fit with a phenomenological gaussian broadening (left panel).

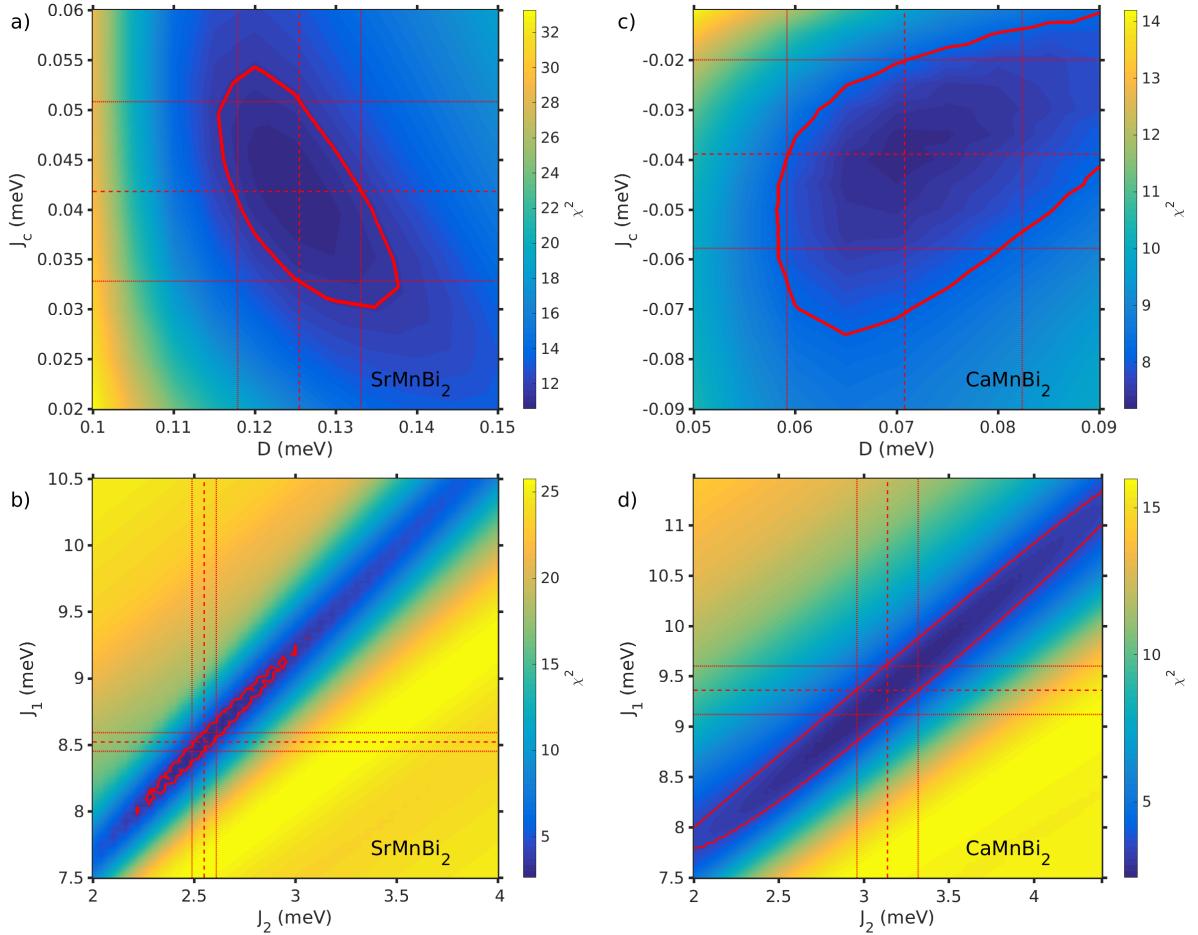


Fig. S8 (color online). χ^2 maps of least-squares fits of the linear spin-wave model for SrMnBi₂ (a,b) and CaMnBi₂ (c,d). Panels (a) and (c) reflect fits of a low energy ($E \leq 20$ meV) cut along the $(10L)$ direction of reciprocal space, using the resolution properties calculated by the RESTRAX ray-tracing routine^{20,21} (best-fit result shown in Fig. 6 of the main article). Panels (b) and (d) illustrate fit results of the high-energy ($30 \leq E \leq 45$ meV) dispersion in the $(HK0)$ plane, used to determine J_1 and J_2 . The red lines represent the one- σ contour. Dashed lines indicate the best-fit values and error margins quoted in the main article. Note that while J_1 and J_2 appear to be strongly correlated, weak details of the dispersion in the $(HK0)$ plane are not adequately reflected in the χ^2 value and constrain the values further than suggested by the one- σ contour.