Formation of a topological non-Fermi liquid in MnSi

R. Ritz1, M. Halder1, M. Wagner1, C. Franz1, A. Bauer1 & C. Pfleiderer1

Fermi liquid theory provides a remarkably powerful framework for the description of the conduction electrons in metals and their ordering phenomena, such as superconductivity, ferromagnetism, and spin- and charge-density-wave order. A different class of ordering phenomena of great interest concerns spin configurations that are topologically protected, that is, their topology can be destroyed only by forcing the average magnetization locally to zero1. Examples of such configurations are hedgehogs (points at which all spins are either pointing inwards or outwards) and vortices. A central question concerns the nature of the metallic state in the presence of such topologically distinct spin textures. Here we report a high-pressure study of the metallic state at the border of the skyrmion lattice in MnSi, which represents a new form of magnetic order comprised of topologically non-trivial vortices2. When long-range magnetic order is suppressed under pressure, the key characteristic of the skyrmion lattice—that is, the topological Hall signal due to the emergent magnetic flux associated with the topological winding—is unaffected in sign or magnitude and becomes an important characteristic of the metallic state. The regime of the topological Hall signal in temperature, pressure and magnetic field coincides thereby with the exceptionally extended regime of a pronounced non-Fermi-liquid resistivity3,4. The observation of this topological Hall signal in the regime of the NFL resistivity suggests empirically that spin correlations with non-trivial topological character may drive a breakdown of Fermi liquid theory in pure metals.

To address the nature of the metallic state at the border of long-range topological order, we have selected the B20 compound MnSi. At zero pressure, $p = 0$, MnSi undergoes a fluctuation-induced first-order transition to helimagnetic order at $T_c = 29.5\,K$ (ref. 4). The helimagnetism originates in a hierarchy of three energy scales, comprising ferromagnetic exchange on the strongest scale, Dzyaloshinsky–Moriya interactions on an intermediate scale and higher-order spin–orbit coupling on the weakest scale5. As a function of magnetic field, $B$, conical order appears for $B > B_c^I \approx 0.1\,T$, and this is followed by a spin-polarized state for $B > B_c^S \approx 0.6\,T$. A phase pocket in the vicinity of $T_c$, known as the A-phase, supports the skyrmion lattice6. The magnetic phase diagram of MnSi including the skyrmion lattice is thereby generic for all helimagnetic B20 compounds, regardless whether they are high-purity metals7, semiconductors8,9 or insulators10,11. As a function of pressure, the helimagnetic transition in MnSi vanishes above $p_c \approx 14.6\,kbar$ without quantum criticality12,13. Yet the resistivity changes from the $T^2$ dependence of a Fermi liquid to the $T^{5/2}$ dependence of a non-Fermi-liquid (NFL) when $p > p_c$. The exceptionally wide NFL range5,14 and the lack of sample dependence of the $T^{5/2}$ coefficient contrast with the excellent quantitative description of MnSi as a weak itinerant magnet15, suggesting that the cause of the NFL behaviour may be an intrinsic mechanism mimicking the effects of disorder and glassiness. This notion is supported by neutron scattering, NMR and muon spin resonance measurements, suggesting partial magnetic order on timescales between $10^{-16}\,s$ and $10^{-11}\,s$ (refs 11, 13). In turn, several theoretical studies15–17 have explored a proliferation of topological spin textures as the cause of the partial order, with a possible link to the NFL resistivity18. However, until now, evidence for topologically non-trivial spin textures in the NFL regime as well as a link between such textures and the NFL behaviour has not been reported.

A unique experimental probe of the topology of magnetically ordered states is the Hall effect, which reflects the Berry phases developed by the electrons as they follow the spin orientation of the magnetic structure. The consequences of these Berry phases may be viewed in terms of an emergent magnetic field with two limiting types of behaviour19. For variations on atomic scales, the emergent field acts essentially in reciprocal space, giving rise to dissipationless Hall currents; this is referred to as the intrinsic anomalous Hall effect. In contrast, for smooth variations on length scales much larger than the Fermi wavelength, the emergent field acts similarly to a real magnetic field, giving rise to a topological Hall signal. Experimentally, these two limits may be readily distinguished in terms of their dependence on temperature and magnetization. Namely, the Hall resistivity associated with the intrinsic anomalous Hall contribution scales with the square of the longitudinal resistivity, $\rho_{xx}$. Therefore, in ‘good’ metals, where $\rho_{xx}$ rapidly decreases with temperature, the intrinsic anomalous contribution to $\rho_{xy}$ also decreases with decreasing temperature. This difference from the topological contribution to $\rho_{xy}$, which increases rapidly with decreasing temperature, owing to the increase in the spin polarization and a reduction in spin-flip scattering19.

At ambient pressure, the Hall effect in MnSi is dominated by the sum of a normal contribution and an intrinsic anomalous contribution20. In addition, a topological Hall signal has been observed in the skyrmion lattice phase17,21. Moreover, a giant topological Hall signal exists for pressures between 6 and 12 kbar (ref. 22). However, that study22 failed to connect the giant topological Hall signal experimentally with the skyrmion lattice phase at ambient pressure and the NFL behaviour at high pressure. In addition, the size and field range of the Hall signal seemed anomalously large and at odds with the behaviour expected at ambient pressure.

To search for a link between the skyrmion lattice and the NFL resistivity, and to resolve the origin of the giant topological Hall signal reported in ref. 22, we performed an extensive high-pressure study exploring the roles of sample purity, pressure transmitter, cooling procedure and sample orientation. To this end, we assembled eight pressure cells covering nearly 40 pressure points (details of the experimental methods are reported in ref. 19 and Supplementary Information). As an important first step in studying the complex phase diagram near $p_c$, we demonstrated that the giant topological Hall signal at low pressures reported in ref. 22 is connected to the skyrmion lattice phase at $p = 0$. We found that the large size of the signal is intrinsic, and that the large field range is most probably due to low sample purity and inhomogeneous pressure conditions.

Here we report that the topological Hall signal of the skyrmion lattice evolves into a prominent characteristic of the entire NFL regime for pressures up to 18.1 kbar, the highest pressure we investigated, which is much greater than $p_c$. Our conclusions are based on the sign and size of the topological Hall signal and the observation of clear boundaries of this signal (Fig. 1a, b) that reproduce the boundaries of the NFL behaviour of $\rho_{xx}$ as a function of pressure, magnetic field and temperature3,11,12. First, for pressures exceeding $p^* \approx 12\,kbar$ there is
a clear crossover with decreasing temperature from the regime at high temperatures to a NFL resistivity at $T^* \approx 12$ K, and $\rho_{xx}$ settles into a stable $T^{3/2}$ NFL dependence at temperatures below $\sim 8$ K. In fact, for what follows below it is essential to emphasize that for $T_c \lesssim T^*$, the crossover and associated NFL behaviour emerges between $T_c$ and $\sim T^*$ (ref. 3; below $T_c$, Fermi liquid behaviour is observed). Second, as mentioned above, the dependence of $\rho_{xx}$ on $T$, $\rho_{xx}(T) \propto T^\alpha$, changes abruptly at $p_c$ from a Fermi liquid form ($\alpha = 2$), to the NFL form ($\alpha = 3/2$) (Fig. 1a, inset). Third, for $p > p_c$, the NFL behaviour returns abruptly as a function of field to Fermi liquid behaviour at a value, $B_{NFL}$, that coincides with the itinerant metamagnetic transition, at $B_{in}$ (Fig. 1b, inset; we refrain here and in Fig. 2b from showing exponents in the centre of the crossover very close to $B_{NFL}$.

To be able to present our main observations further below, which requires distinguishing topological contributions to the Hall signal from those that are anomalous, we show in Fig. 1c, d, our magnetotransport data over a large field range, $-14$ to $14$ T. At $2.8$ K and at low pressures, $\rho_{xy}$ drops by $\sim 10\%$ at $B_{G2}$, and this is followed by a gradual increase towards high fields (Fig. 1c, upper panel). Above $T_c$, this decrease in $\rho_{xx}$ broadens and shifts to higher fields (Fig. 1c, lower panel). For $p > p_c$, $\rho_{xx}$ displays a similar decrease at $B_{in}$ (the itinerant metamagnetic transition) with a temperature dependence for $B_{in}$ that is empirically similar to what is seen above $T_c$ for low pressures, that is, $B_{in}$ shifts to higher values. This is consistent with spin scattering processes in the NFL regime that are quenched above $B_{NFL}$. Furthermore,

---

**Figure 1** | Temperature and field dependence of the resistivity, $\rho_{xx}$, and Hall resistivity, $\rho_{xy}$, of MnSi over a wide range. Data shown here were measured in pressure cell pc8 (see also ref. 19 and Supplementary Information). a, $\rho_{xx}$ at various pressures (see also refs 29). Inset, exponent $x$ inferred from $\rho_{xx}$ (see also refs 3, 30). b, $\rho_{xx}$ at various fields for $p = 15.7$ kbar. Inset, exponent $x$ at $p = 15.7$ kbar. c, $\rho_{xy}$ at 2.8 K and 28.7 K up to 15.7 kbar. d, $\rho_{xy}$ up to 15.7 kbar. At 28.7 K a strong knee-shaped intrinsic anomalous Hall contribution emerges; that is, an additional shoulder emerges on top of the intrinsic anomalous Hall contribution that merges with the itinerant metamagnetism above $p_c$ and at low temperatures. FL, Fermi liquid; NFL, non-Fermi liquid.

---

**Figure 2** | Hall resistivity, $\rho_{xy}$, and magnetoresistance, $\rho_{xx}$, for low fields at various pressures. a, $\rho_{xy}$ for a wide range of pressures and temperatures. b, Comparison of the magnetoresistance, $-\rho_{xx}$; the Hall resistivity after subtraction of normal contributions, $\rho_{xy} - \rho_{xy,normal}$ and the exponent of the temperature dependence of the electrical resistivity, $\alpha$, at various temperatures, for a pressure greater than $p_c$. With increasing field, $\alpha$ changes from NFL behaviour ($\alpha = 3/2$) to Fermi liquid behaviour ($\alpha = 2$) at the same field value, above which the topological Hall contribution has vanished. PM, paramagnet; pc2 and pc8 refer to the pressure cells studied (Supplementary Information).
the a.c. susceptibility\textsuperscript{19,23}. A discussion of the associated temperature dependences is presented in Supplementary Information. Hence, for large fields $\rho_{xx}$ is dominated by the sum of a normal contribution and an intrinsic anomalous contribution\textsuperscript{20}, of which the latter displays an additional shoulder (see also Fig. 3). The shoulder in $\rho_{xx}$ can therefore be accounted for neither by variations in $\rho_{xx}$ (entering the Hall conductivity $\sigma_{xy} = -\rho_{xy}/(\rho_{xx}^2 + \rho_{xx}^2)$, which is proportional to the intrinsic anomalous Hall effect) nor by the magnetization, the field dependence of which is qualitatively similar above $T_c$ for all pressures, with no signs of a shoulder\textsuperscript{19,23}. This suggests a more subtle connection between the shoulder in $\rho_{xx}$ and the itinerant metamagnetism, but this connection is not important for the conclusions of our study.

Typical data highlighting the topological contributions to the Hall signal for samples with high residual resistivity ratios (RRRs) are summarized in Fig. 2. Qualitatively similar data for samples with low RRRs are presented in Supplementary Information. To illustrate the relationship between the topological Hall signal and the nature of the metallic state inferred from the temperature dependences of $\rho_{xx}$, we use the following colour scheme: white for the paramagnetic regime, sky blue for the Fermi liquid and orange for the NFL. For low pressures, a topological Hall signal arises exclusively in the A-phase\textsuperscript{19} (red shading). It is important to emphasize that the change in slope of $\rho_{xx}$ in this regime (for example at 0.5 T for 7 kbar and 16.6 K) is due to the conical-to-ferromagnetic transition at $B_{c2}$ and is therefore completely unrelated to the itinerant metamagnetism at $B_S$ at high pressures and above $T_c$.

As $T_c$ decreases below $T^* \approx 12$ K, the topological signal is also present above $T_c$ up to $-T^*$ and over a wide field range where the magnitude and sign of the signal are unchanged compared with the data below $T_c$ (see data for 13.7 kbar in Fig. 2a). Because the shoulder in the itinerant metamagnetism data at these temperatures and pressures is located at a field strength of around several teslas (well outside the field range shown here; see Supplementary Fig. 4), this unambiguously connects the topological Hall signal in the A-phase with the NFL resistivity. At pressures greater than $p_c$, the topological contribution is observed across the entire NFL regime. We have confirmed this observation in several samples of differing purity, where the topological Hall contribution tends to be larger and broadened for these samples with lower RRRs. We thereby note that, regardless of the RRR, there are no hints of metastable behaviour such as that observed under field cooling at low pressures (see Supplementary Fig. 6 and fig. 7 in ref. 19).

To justify the positioning of regime boundaries in Fig. 2a, we show in Fig. 2b, for $p = 14.7$ kbar $> p_c$, a typical comparison of $\rho_{xx}$ with the Hall resistivity after subtracting the normal Hall contributions, $\rho_{xy} = \rho_{xy,norm} - \rho_{xy,norm}$ (where $\rho_{xy,norm}$ was inferred from data above $B_{c2}$), and with the exponent, $\alpha$, of the temperature dependence of the resistivity at selected fixed fields. As a function of field, the following characteristic features coincide at $B_{NFL}$: the onset of the decrease in the magnetoresistance suggestive of a field-induced suppression of a magnetic scattering mechanism, the disappearance of the topological Hall contribution (Fig. 2b, red shading) and the change from Fermi liquid to NFL resistivity. We can therefore confirm that the field dependence of $\rho_{xx}$ neither qualitatively nor quantitatively causes variations in the intrinsic anomalous Hall contribution ($\sigma_{xy} \approx -\rho_{xy}/\rho_{xx}^2$) that may be taken for a topological Hall signal. Thus, the topological Hall signal and the NFL resistivity coincide as functions of $T$, $B$ and $p$.

In Fig. 3, we show the relevant regions of the pressure–temperature phase diagram and typical magnetic phase diagrams. For zero field, NFL behaviour emerges below $T^*$ and above $T_c$ as well as below $T^*$ for pressures greater than $p_c$. The NFL behaviour is accompanied by a topological Hall signal that is present even for very low fields, as marked by NFL + TH in each panel. For pressures below $p^* \approx 12$ kbar, the magnetic phase diagram remains qualitatively unchanged as compared with ambient pressure (Fig. 3b), and the topological Hall signal is enhanced as reported in ref. 19. Qualitatively new behaviour emerges for $p > p^*$, where the topological Hall contribution survives above $T_c$ and $p_c$. This is illustrated in Fig. 3c, d, e, where the boundaries of the regime of the NFL resistivity are shown as a function of $T$, $B$ and $p$.

The sign and the magnitude of the topological Hall signal arises from a combination of the strength of the emergent field, given by the topological winding number for the magnetic unit cell; the (local) spin polarization of the conduction electrons; and an average over individual bands taking into account different scattering processes\textsuperscript{19}. The

**Figure 3** | Phase diagrams of MnSi. **a**, Temperature–pressure phase diagram. NFL + TH refers to the regime of an NFL resistivity in which a small field establishes a topological Hall (TH) signal. **b**, Magnetic phase diagram at $p = 7.4$ kbar, displaying a strongly enhanced topological Hall signal in the A-phase as reported in ref. 19. **c**, Magnetic phase diagram at $T_c$ and $p = 13.7$ kbar. The transition at $T_c$ is first order, and the onset of NFL resistivity occurs at a temperature above $T_c$. A topological Hall contribution is observed in the A-phase and the NFL regime above $T_c$. **d, e**, Magnetic phase diagrams at $p = 15.7$ and 18.1 kbar, above $p_c$. The field and temperature range of the topological Hall signal extends over the entire field and temperature range of the NFL resistivity. See Supplementary Information for illustrated definitions of all characteristic fields and temperatures.
observation of neutron scattering intensity above $p_c$ at a wavelength which corresponds to that of the helical state at low pressures implies that the continuous evolution of the topological Hall signal from the A-phase to the NFL regime must be closely connected to an unchanged topological winding number. As a caveat, neutron intensity is observed only in a small region of the NFL regime, whereas the topological Hall signal we report here is seen everywhere. Thus, the topological winding must be insensitive against fluctuations above $p_c$, at least on timescales relevant to the Hall effect. As a consequence, any of the following scenarios (or combinations thereof) can arise in the high-pressure state of MnSi: spontaneous formation of randomly oriented skyrmions at $B = 0$ that are stratified in a magnetic field; formation of skyrmions even at very low fields; strong fluctuations between helical modulations and skyrmionic textures. It therefore seems likely that strong quantum fluctuations promote this state.

Our study identifies a topological Hall signal as a prominent characteristic of the NFL regime of MnSi. The sign and magnitude of the Hall signal indicate that the topological winding of skyrmions as seen at $p = 0$ is the long-sought intrinsic mechanism causing the $T^{3/2}$ NFL dependence. This dependence is characteristic of a strong divergence of the quasiparticle self-energy as $T \to 0$, which suggests a generic breakdown of Fermi liquid theory due to spin correlations with topologically non-trivial character. Notably, several theoretical studies suggest various mechanisms leading to the formation of non-trivial spin textures at the zero-temperature border of itinerant ferromagnetism for different reasons. In fact, experimentally a $T^{3/2}$ NFL resistivity has also been observed near putative ferromagnetic quantum phase transitions, for example in ZrZn$_2$ and Ni$_3$Al. However, if this is related to complex spin textures, their average topological winding would be degenerate and no topological Hall effect would therefore be expected. Hence, the topological character of the NFL regime we report here for MnSi may turn out to be the first example of a more general phenomenon, in which the full suppression of the magnetization is generically preceded by the formation of complex spin textures. For the case of MnSi, the peculiar stability of the NFL behaviour may thereby be inherited from the chiral character of the Dyazholsky–Moriya spin–orbit interaction.

METHODS SUMMARY

Sample preparation. Single crystals of MnSi were grown by optical float zoning under ultrahigh-vacuum-compatible conditions. The specific heat, susceptibility and resistivity of small pieces taken from these single crystals were in excellent agreement with established results. Samples with different RRRs were studied as summarized in Supplementary Information. Samples for the measurements reported here were oriented by Laue X-ray diffraction, cut with a wire saw and carefully polished to size. Current leads were soldered to the small faces of the sample, and platinum wires for the voltage pick-up were spot-welded onto the surface of the sample.

Magnetotransport under pressure. The Hall resistivity and longitudinal resistivity were measured simultaneously using a standard six-terminal phase-sensitive detection system. The voltage signals were amplified with impedance-matching transformers to optimize the signal-to-noise ratio. Low excitation frequencies and excitation currents were applied to minimize parasitic signal pick-up. High hydrostatic pressures were generated with a Cu:Be clamp cell using various different excitation currents. High hydrostatic pressures were measured simultaneously using a standard six-terminal phase-sensitive detector. Magnetotransport under pressure.

Received 3 December 2012; accepted 15 February 2013.

Published online 1 May 2013.

1. Chaspin, P. & Lubensky, T. Principles of Condensed Matter Physics Chs 9, 10 (Cambridge Univ. Press, 1995).

2. Mühlbauer, S. et al. Skyrmion lattice in a chiral magnet. Science 323, 915–919 (2009).

3. Pfleiderer, C., Julian, S. R. & Lonzarich, G. G. Non-Fermi liquid nature of the normal state of itinerant-electron ferromagnets. Nature 414, 427–430 (2001).

4. Janoschek, M. et al. Fluctuation-induced first-order phase transition in Dzyaloshinskii-Moriya helimagnets. Phys. Rev. B (in the press); preprint available at http://arxiv.org/abs/1205.4780 (2012).

5. Landau, L. D. & Lifshitz, E. M. Course of Theoretical Physics Vol. 8, Sec. 52 (Pergamon, 1980).

6. Xu, Z. et al. Real-space observation of a two-dimensional skyrmion crystal. Nature 465, 901–904 (2010).

7. Yu, Z. et al. Near-room-temperature formation of a skyrmion crystal in thin-films of the helimagnet FeGe. Nature Mater. 10, 106–109 (2011).

8. Muller, W. et al. Skyrmion lattice in the doped semiconductor Fe$_3$CoSi. Phys. Rev. B 81, 041203 (2010).

9. Seki, S., Yu, X., Ishiwata, S. & Tokura, Y. Observation of skyrmions in a multiferroic material. Science 336, 198–201 (2012).

10. Adams, T. et al. Long-wavelength helimagnetic order and skyrmion lattice phase in Cu$_2$OSeO$_3$. Phys. Rev. Lett. 108, 237204 (2012).

11. Pfleiderer, C. et al. Partial order in the non-Fermi liquid phase of MnSi. Nature 427, 227–231 (2004).

12. Pfleiderer, C., Böni, P., Keller, T., Rößler, U. & Rosch, A. Non-Fermi liquid metal without quantum criticality. Science 316, 1871–1874 (2007).

13. Uemura, Y. et al. Phase separation and suppression of critical dynamics at quantum transitions of itinerant magnets: MnSi and (Sr$_x$Ca$_{1-x}$)$_3$Ru$_2$O$_{12}$. Nature 447, 3–9 (2007).

14. Lonzarich, G. G. & Taillefer, L. Effect of spin fluctuations on the magnetic equation of state of ferromagnetic or nearly ferromagnetic metals. J. Phys. C 18, 4339–4371 (1985).

15. Tewari, S., Belitz, D. & Kirkpatrick, T. R. Blue quantum fog: chiral condensation in quantum helimagnets. Phys. Rev. Lett. 96, 047207 (2006).

16. Binz, B., Vishwanath, A. & Ayi, N. Theory of the helical spin crystal: a candidate for the partially ordered state of MnSi. Phys. Rev. Lett. 96, 207202 (2006).

17. Rößler, U. K., Bogganov, A. N. & Pfleiderer, C. Spontaneous skyrmion ground states in magnetic metals. Nature 442, 797–801 (2006).

18. Kirkpatrick, T. R. & Belitz, D. Columnar fluctuations as a source of non-Fermi-liquid behavior in weak metallic magnets. Phys. Rev. Lett. 104, 236404 (2010).

19. Ritz, R. et al. Giant generic topological Hall resistivity in MnSi under pressure. Phys. Rev. B (submitted).

20. Lee, M., Onose, Y., Tokura, Y. & Ong, N. P. Hidden constant in the anomalous Hall effect of high-purity magnet MnSi. Phys. Rev. B 75, 172403 (2007).

21. Neubauer, A. et al. Topological Hall effect in the A phase of MnSi. Phys. Rev. Lett. 102, 186602 (2009).

22. Lee, M., Kang, W., Onose, Y., Tokura, Y. & Ong, N. Unusual Hall effect anomaly in MnSi under pressure. Phys. Rev. Lett. 102, 186601 (2009).

23. Thessieu, C., Pfleiderer, C., Stepanov, A. N. & Fleouf, J. Field dependence of the magnetic quantum phase transition in MnSi. J. Phys. Condens. Matter 9, 6677–6687 (1997).

24. Pfleiderer, C., Reznik, D., Pintschovius, L. & Haug, J. Magnetic field and pressure dependence of small angle neutron scattering in MnSi. Phys. Rev. Lett. 93, 156406 (2007).

25. Belitz, D. & Kirkpatrick, T. R. Fluctuation-driven quantum phase transitions in clean itinerant ferromagnets. Phys. Rev. Lett. 89, 247202 (2002).

26. Conduit, G. J., Green, A. G. & Simons, B. D. Inhomogeneous phase formation on the border of itinerant ferromagnetism. Phys. Rev. Lett. 103, 207201 (2009).

27. Smith, R. P. et al. Marginal breakdown of the Fermi-liquid state on the border of metallic ferromagnetism. Nature 455, 1220–1223 (2008).

28. Neubauer, A. et al. Ultra-high vacuum compatible image furnace. Rev. Sci. Inst. 82, 013902 (2011).

29. Pfleiderer, C., McMullan, G. J., Julian, S. R. & Lonzarich, G. G. Magnetic quantum phase transition in MnSi under hydrostatic pressure. Phys. Rev. B 85, 8330–8338 (2017).

30. Doiron-Leyraud, N. et al. Fermi-liquid breakdown in the paramagnetic phase of a pure metal. Nature 425, 595–599 (2003).

Supplementary Information is available in the online version of the paper. Acknowledgements We wish to thank P. Böni, K. Everschor, M. Garst, M. Janoschek, S. Mayr and A. Rosch for discussions and support; R.R., M.H., A.B., M.W. and C.F. acknowledge financial support within the TUM Graduate School. Financial support through DFG TRR80 and DFG FOR960 as well as ERC-Ag (291079 TOPFIT) are gratefully acknowledged. Author Contributions R.R. and C.P. developed the experimental set-up; R.R. performed the transport measurements; M.H. and M.W. performed magnetization measurements; C.F. wrote the software for analysing the data; A.B. grew the single-crystal samples and characterized them; R.R. and C.P. analysed the experimental data; C.P. supervised the experimental work; C.P. proposed this study and wrote the manuscript; all authors discussed the data and commented on the manuscript.

Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to R.R. (robert.rizzi@frm2.tum.de) or C.P. (christian.pfleiderer@frm2.tum.de).