Reexamination of the microscopic couplings of the quasi one–dimensional antiferromagnet CuGeO$_3$

K. Fabricius, A. Klümper, U. Löw  
Fachbereich Physik, Bergische Universität Wuppertal, Gaußstr. 20, 42097 Wuppertal, Germany  
Institut für Theoretische Physik, Universität zu Köln, Zülpicher Str. 77, 50937 Köln, Germany  
Institut für Physik, Universität Frankfurt, Robert-Mayer-Str. 2-4, 60325 Frankfurt, Germany

B. Büchner, T. Lorenz  
II. Physikalisches Institut, Universität zu Köln, Zülpicher Str. 77, 50937 Köln, Germany  
(March 23, 2022)

Abstract

Experimental data for the magnetic susceptibility and magnetostriction of CuGeO$_3$ are analyzed within a one-dimensional antiferromagnetic model with nearest ($J_1$) and next-nearest neighbour interactions ($J_2$). We show that the ratio of the exchange constants in the antiferromagnetic chains of CuGeO$_3$ amounts to $J_2/J_1 = 0.354(0.01)$, i.e. it is significantly larger than the critical value for the formation of a spontaneous gap in the magnetic excitation spectrum without lattice dimerization. The susceptibility data are reproduced by our numerical results over the temperature range from 20K to 950K to a high degree of accuracy for $J_1 = 80.2(3.0)$ and $J_2 = 28.4(1.8)$. The pressure dependence of the exchange constants is estimated from magnetostriction data. Furthermore, the specific heat data are checked on consistency against the calculated entropy of the above model.

PACS: 75.80.+q, 75.50.Ee, 75.40 Cx, 75.10.Jm
I. INTRODUCTION

The investigation of low dimensional quantum spin systems has attracted widespread and general interest in active research on a large class of magnetic materials, both experimentally as well as theoretically, since the properties are strongly affected by quantum fluctuations. In particular, there is strong theoretical and experimental effort to understand the origin of singlet-triplet spin gaps in low dimensional spin systems such as $SrCu_2O_3$ and $VO_2P_2O_7$, which occur in the presence of competing spin–interactions, e.g. due to a spin ladder geometry.

Evidence for frustrated spin–interactions has also been reported for the quasi–one–dimensional antiferromagnet $CuGeO_3$. Initially this compound has received much interest, since it is the first example of an inorganic compound undergoing a spin–Peierls transition. Subsequent extensive experimental studies have revealed that many properties of the ordered phase are well described by the well known spin–Peierls scenario, such as the presence of a lattice dimerization and a singlet triplet spin–gap scaling with the lattice distortion. However, recently it was found that the formation of the non–magnetic low temperature phase in $CuGeO_3$ sensitively depends on details of the magnetic exchange.

The traditional spin–Peierls theory is based on one–dimensional antiferromagnetic chains with nearest-neighbour couplings only. Such magnetic systems do not show any long-range antiferromagnetic order in the groundstate, however they possess critical quantum fluctuations which drive the lattice dimerization, i.e. the spin-Peierls transition. Substances with frustrated spin interactions are not strictly governed by as the magnetic system in these cases may show spontaneous long-range magnetic dimerization in the groundstate even without any lattice dimerization. The additional spin-phonon coupling merely stabilizes this magnetic dimerization upon developing the lattice distortion. Whether this or the former scenario is realized depends on the strength of the frustration parameter $\alpha = J_2/J_1$.

Frustration of the spin–interactions in $CuGeO_3$ has been inferred previously from the investigation of the magnetic susceptibility ($\chi$) in the non-dimerized phase, which is in disagreement with a nearest neighbour Heisenberg model. A much better agreement has been found in theoretical studies of the spin–susceptibility $\chi(T)$ invoking a Heisenberg chain with competing nearest and next-nearest neighbour exchange couplings $J_1$ and $J_2$.

However, two markedly different choices of exchange couplings, i.e. $(J_1, \alpha) = (75K, 0.24)$ and $(J_1, \alpha) = (80K, 0.36)$, were derived within model calculations and the same experimental data for the magnetic susceptibility as well as inelastic neutron scattering. For these quantities an increase of $\alpha$ on one hand and a decrease of $J_1$ on the other have similar consequences leading apparently to a large uncertainty of the exchange parameters.

Thus these previous studies of the quantum magnetism in $CuGeO_3$ reveal some evidence for the relevance of magnetic frustration in $CuGeO_3$, whereas it is obviously difficult to extract precise values of the exchange parameters. A precise knowledge of the ratio $\alpha$ is of course very important, since the theory predicts a critical ratio $\alpha_c$ for a spin–gap to develop in the magnetic excitation spectrum. This gap opens irrespective of a lattice distortion. The existence of this gap is established exactly at the Majumdar–Ghosh point $\alpha = 1/2$ and
by several numerical studies \cite{12, 2, 1} which strongly suggest $\alpha_c \approx 0.2411$.

In order to decide whether the spin gap in CuGeO$_3$ is at least partially a manifestation of frustrated spin–interactions in a low dimensional magnet we have performed a comparative study of theoretical results and experimental data for thermodynamic properties. Using our numerical results for the Heisenberg chain with nearest and next-nearest neighbour interaction we show that a comparison of the magnetic susceptibility alone allows for the unique determination of the exchange constants in the frustrated one-dimensional magnet CuGeO$_3$ with result $\alpha = 0.354 \pm 0.01$, i.e. a frustration significantly larger than the critical value $\alpha_c$.

In the course of our investigations we also determine the pressure dependence of the coupling parameters from magnetostriction data. We also analyse the experimental specific heat data on consistency with the theoretical entropy results.

**II. THEORY AND NUMERICS**

The dominant magnetic interactions in CuGeO$_3$ are due to Heisenberg spin exchange between Cu$^{2+}$ ions along the c-axis of the crystal. The Hamiltonian for the spin chain in the non-dimerized phase reads

$$H = 2 \sum_i (J_1 S_i S_{i+1} + J_2 S_i S_{i+2}),$$

where we have adopted the normalization factor of \cite{13}. The nearest neighbour coupling $J_1$ is induced by the exchange path Cu – O – Cu, and the next-nearest neighbour coupling $J_2$ is caused by the path Cu – O – O – Cu. A microscopic calculation of $J_{1,2}$ is difficult \cite{14} and independent derivations are important.

In order to obtain quantitative results we calculate various physical properties in dependence on the couplings $J_1$ and $J_2$, notably the magnetic susceptibility, and perform a two-parameter fit of the experimental data which have been measured up to 950K. The aim is to achieve a best fit above the transition temperature $T_{SP} = 14.3K$ within the model of a magnetic system with interactions described above and an adiabatic decoupling of the spin-phonon interactions. In this sense $J_1$ and $J_2$ are treated as effective coefficients explicitly dependent on the lattice geometry, i.e. microscopic bond angles and lattice constants. Unfortunately, analytic results for the thermodynamics of the model are available only for $\alpha = 0$ (nearest-neighbour Heisenberg chain). We therefore resort to complete numerical diagonalizations of finite systems with chain lengths up to $L = 18$. In general the numerical treatment of strongly correlated quantum spin chains is plagued by finite-size effects at low temperatures. Here, however, we are interested in relatively high temperatures with $k_B T > 0.5 J_1$. A comparison of numerical data for successive chain lengths $L = 16, 17, 18$ shows that finite size corrections are essentially negligeble for our purposes. In Figs. 1 and 2 numerical results for the magnetic susceptibility and specific heat per lattice site are depicted. Note the characteristic dependence of the extremal values $\chi_{max}$ and the corresponding $T_{max}$ on the frustration parameter $\alpha$. $T_{max}$ is decreasing with increasing $\alpha$, whereas $\chi_{max}$ is increasing. The behaviour of the specific heat is similar, however its maximal value $C_{max}$ is a decreasing function of $\alpha$. Consequently, at low temperatures the entropy strongly increases with increasing $\alpha$. 

3
We first observe that the experimental susceptibility data $\chi$ allow for an unambiguous determination of $J_1$ and $J_2$ if we use the position $T_{\text{max}} = 56$ K and absolute value $\chi(T_{\text{max}})$ of the maximum of $\chi$. In practice, we determine for a sequence of frustration parameters $\alpha = J_2/J_1$ the value of $J_1$ leading to $T_{\text{max}} = 56$ K. For this set of coupling parameters the value of $\chi_{\text{max}}$ is calculated, see Fig. 4. The experimental value for $\chi_{\text{max}}$ is obtained in this plot for $\alpha = 0.354$ within an error of 0.01. Note that $\alpha = 0.24$ as used in [2] would yield a value of $\chi_{\text{max}}$ far too large in comparison with the actually measured value. The nearest neighbour coupling corresponding to $\alpha = 0.354$ is $J_1 = 80.2K \pm 3.0$. In Fig. 4 the susceptibility $\chi$ for the values $\alpha = 0.354$ and 0.24 is compared with the experimental data. Note the strong deviation of the theoretical curve for $\alpha = 0.24$ from the measured one especially at $T_{\text{max}}$. In contrast to this, the entire temperature dependence of $\chi(T)$ is reproduced very well for $\alpha = 0.354$. The effect of finite-size corrections on the numerical results has been reduced by a scaling analysis based on a transfer matrix approach [16]. The theoretical results are reliable down to temperatures of 35 K. Note that a Landé factor of $g = 2.256$ has been used as derived from ESR [17]. (Furthermore we have assumed a cancellation of van Vleck paramagnetism and core diamagnetism which is the most reasonable assumption about the magnetic background. If we subtract a background of $5 \cdot 10^{-5}$ emu/mole the result would read $\alpha = 0.362 \pm 0.01$.) For the quoted error bars of $J_1$, $\alpha$ a relative error of 3% in the measurements has been taken into account. Also note that a comparable analysis of the data [4] leads to $\alpha = 0.371 \pm 0.01$ ($\alpha = 0.38 \pm 0.01$ if a background of $5 \cdot 10^{-5}$ emu/mole is subtracted), a value which overlaps with the above result. The differences between our and the data in ref. [4] might originate from slightly different orientations of the magnetic fields with respect to the crystal axes. Note that our data agree much better with those in ref. [13].

In addition to the strength of the microscopic couplings we can determine their pressure dependence. We obtain this from magnetostriction data [3] which are related to the pressure dependence of the magnetic susceptibility [18-21] which is rather directly accessible in numerical studies

$$\frac{1}{L_i} \left( \frac{\partial L_i}{\partial H} \right)_{p_i} = \frac{H}{V} \left( \frac{\partial \chi}{\partial p_i} \right) _H = \frac{H}{V} \left( \partial_1 \chi \left( \frac{\partial J_1}{\partial p_i} \right)_T + \partial_2 \chi \left( \frac{\partial \chi}{\partial p_i} \right)_T \right), \quad (2.2)$$

where $\partial_{1,2}$ denotes the derivative of $\chi$ with respect to $J_1$ and $\alpha$. Based on this relation and the data for temperatures 40 K and 60 K we find $\partial J_1/\partial p_i = 3.3(3), -7.5(5), -1.4(2)$ K GPa$^{-1}$, and $\partial \alpha/\partial p_i = -0.03(3), 0.01(4), 0.04(2)$ GPa$^{-1}$ for the three lattice axes $i = a$, $b$, and $c$. Note that the values for the pressure dependence of $\alpha$ obtained along this way are consistent with the hydrostatic pressure dependence obtained in [20]. However, in our analysis the hydrostatic pressure dependence of $J_1$ is much stronger than that of $J_2$ which is essentially zero.

III. ENTROPY ANALYSIS

Unfortunately it is impossible to measure the temperature dependence of the magnetic specific heat in CuGeO$_3$ directly. As displayed in the left part of Fig. 4 at temperatures close to the predicted maximum of $C_{\text{mag}}$ the total specific heat is dominated by the phonon contribution $C_{\text{ph}}$, i.e. it is about one order of magnitude larger than the calculated magnetic
contribution. Therefore it is impossible to extract $C_{mag}$ with sufficient accuracy to resolve the small differences predicted for different exchange constants in a $J_1 - J_2$ model, see Fig.2. Note that this would require a knowledge of the phonon contribution with an unrealistic accuracy of the order of $10^{-3}$ or higher. Nevertheless, one can use the measurements of $C$ to check different exchange parameters suggested in the literature \[11,24,26\] for consistency. In the following we will demonstrate that indeed most of these values for $J_1, J_2$ lead to discrepancies.

It is well known that a reliable separation of magnetic and phonon contributions of $C$ is possible at low temperatures \[22,23,27\] and we will use this separation to estimate the magnetic entropy at higher temperatures. Note that the spin–Peierls phase transition does not modify $C_{ph}$ significantly, i.e. the anomaly of $C$ is due to the magnetic contribution. This is inferred on one hand from the extremely small structural changes at the phase transition and on the other hand consistent with the findings in measurements as functions of magnetic fields and doping \[21,27\].

Analyses of the specific heat at low temperatures have been reported several times \[22,23,27\]. Well below $T_{SP}$ $C_{mag}$ shows activated behavior due to the large spin gap. Moreover, at low temperatures, i.e. at and below $T_{SP}$, the specific heat and correspondingly the entropy are dominated by their magnetic contributions. At low temperatures the phonon contribution follows the usual $T^3$ law, i.e. $C_{ph} = \beta T^3$ with $\beta \simeq 0.3 mJ/K^4mole$. At higher temperatures $C_{ph}$ deviates from this $T^3$ behavior as illustrated in Fig.5. The extrapolation of the low temperature behavior exceeds the total specific heat already at moderate temperatures of about 30K. Therefore it may serve as an upper limit for $C_{ph}$, which we use to derive a lower limit $C_{mag}^{min}$ for the magnetic contribution from the data.

Below 20K we determine this lower limit simply from the difference $C - \beta T^3$ by assuming a large value of $\beta \simeq 0.32 mJ/K^4mole$. The difference function shows a maximum well above $T_{SP}$ at 20K. A further extrapolation of this function to higher temperatures leads to a decrease which is unreasonable for an estimate of $C_{mag}$. We therefore take the value of $C - \beta T^3$ at 20K as a lower limit of $C_{mag}$ at temperatures above 20K (see Fig.5). Of course this treatment yields only a very small lower limit for $C_{mag}$, which is indeed much smaller than the predicted $C_{mag}$ (see Fig.5a). We do not aspire a more realistic extraction of $C_{mag}$ here, since on one hand it is impossible to achieve the necessary accuracy to fix the exchange constants and on the other hand the very conservative estimate of $C_{mag}^{min}$ already suffices to rule out certain exchange constants reported for CuGeO$_3$.

¿From the lower limit of $C_{mag}$ we have calculated the corresponding minimum magnetic entropy $S_{mag}^{min}$, which is compared to calculations of $S_{mag}$ for several choices of $J_1$ and $J_2$ in the right part of Fig.5. Taking the temperature at the maximum of $\chi$ and assuming $\alpha = 0$ yields $J = 44K$ as described in the initial paper on the spin–Peierls transition in CuGeO$_3$ \[1\]. The corresponding entropy is much larger than the lower limit we have extracted from the data. Nevertheless, the specific heat data allow to exclude these parameters. At temperatures slightly above $T_{SP}$ the calculated magnetic entropy for $J_1 = 44K$, $J_2 = 0$ amounts to about 95% of the total entropy in CuGeO$_3$. In other words an unreasonably small phonon background, e.g. with a $\beta \simeq 0.04 mJ/moleK^4$ which is nearly one order of magnitude too small, has to be chosen to account for this large magnetic entropy.

A further suggested value for the intrachain exchange constant in a model with $\alpha = 0$ is $J = 60K$. This value has been reported first in \[10\] based on their inelastic neutron
scattering data. More recently in [24] the same value and in addition a large (frustrated) intrachain exchange was found from the analysis of the dispersion curves in the dimerized phase, i.e. below $T_{SP}$. It is apparent from Fig.4 that the magnetic entropy calculated for these exchange parameters in a one-dimensional model, i.e. for $J_1 = 60K$, $J_2 = 0$ and above $T_{SP}$, is significantly smaller than the lower limit $S_{mag}^{min}$ extracted from the data. Thus the specific heat does not support the parameters suggested in ref. [24], in particular when taking into account a further reduction of the theoretical $S_{mag}$ due to the suggested large interchain exchange.

The three solid lines in the right part of Fig.5 correspond to calculations of $S_{mag}$ for different exchange constants and finite $\alpha$. Since these entropies have been calculated for finite chains, the results are reliable for temperatures above approx. 35K.

The strongest discrepancy between numerical and experimental data is present for the set of parameters $J_1 = 125K, \alpha = 0.35$. These parameters have been suggested recently in [25] to give the best description of the magnetic specific heat in $CuGeO_3$. In this latter work $C_{mag}$ has been extracted from Raman scattering data and it was concluded that it is impossible to fit both the susceptibility and the magnetic specific heat with a single choice of $J_1$ and $J_2$. However, it is apparent from Fig.4 that the parameters suggested in ref. [25] and consequently the “magnetic specific heat” extracted from Raman scattering is in striking discrepancy to the measurement of the specific heat at low temperatures. Thus our data do not support the reported inconsistency in the determination of exchange parameters from $\chi$ and $C_{mag}$ [25]. Further investigations seem necessary to explain the striking discrepancy between $C_{mag}$ as revealed in [25] from quasi–elastic scattering and the true magnetic specific heat.

The deviation between the data and the calculations for the exchange constants $J_1 = 75K, \alpha = 0.24$, which have been extracted by Castilla et al. from their analysis of the magnetic susceptibility and the dispersion curves, is less pronounced. The calculation of the magnetic entropy is reliable for temperatures larger than 35K where the numerical results are indeed larger than the lower bound we have estimated from the data. A very nice convergence of theory and experiment appears upon further increasing $\alpha$ to 0.354 (and $J=80.2K$). As shown in Fig.4 the magnetic entropy calculated for the parameters which yield the best fit to the susceptibility is always larger than the lower bound extracted from the data. Moreover, it is apparent from Fig.4 that the difference between the theoretical $S_{mag}(J_1 = 80.2K, \alpha = 0.354)$ and the lower bound systematically decreases with decreasing temperature, i.e. with increasing accuracy of the extracted minimum magnetic entropy.

Unfortunately, it is impossible to extend the theoretical calculations to temperatures of 20K and below. At these temperatures the data for $S_{mag}$ shown in Fig.4 do not only represent a rough lower bound. Close to $T_{SP}$ $S_{mag}$ is markedly larger than $S_{ph}$ for any reasonable $C_{ph}$ (As mentioned above the curve for $S_{mag}(J = 44K)$ corresponds already to 95% of the total entropy). Therefore even assuming a significantly smaller phonon background leads only to moderate changes of $S_{mag}$ at these temperatures.

Boldly extrapolating the calculated entropy for $J = 80.2K, \alpha = 0.354$ to lower temperatures one may conclude that both the value of $S_{mag}$ at $T_{SP}$ as well as its temperature dependence, i.e. the magnetic specific heat, are well described. In contrast to that for all other choices of exchange parameters in Fig.4 significant discrepancies between the model calculations and the experimental data are apparent. Though the accuracy for the deter-
mination of $C_{mag}$ is not sufficient to unambiguously determine the exchange constants from these data alone, the specific heat strongly confirms our analysis of the susceptibility. In particular, in contrast to the conclusions of [25] there is no evidence that it is necessary to invoke markedly different exchange constants to explain $\chi$ and $C_{mag}$ (at low temperatures).

IV. CONCLUSION

We have presented numerical results for thermodynamical properties of a quantum spin-1/2 chain with nearest and next-nearest neighbour interactions which is believed to be at the heart of the magnetic system of CuGeO$_3$. The microscopic interaction parameters have been determined as well as their uniaxial pressure dependence. We have shown that the frustration parameter is $\alpha = 0.354$. This is much larger than the value used for the explanation of Raman scattering data [26,20]. We expect our result to be reliable as we have based our reasoning on established thermodynamical relations. Furthermore, we have demonstrated that the experimental magnetic susceptibility data are accounted for in even quantitative details by the quasi one-dimensional model and $\alpha = 0.354$.

Within the present accuracy the magnetic specific heat calculated for the exchange constants derived from our analysis of $\chi$ is consistent with the analysis of the experimental data. On the other hand for several other choices of exchange parameters which have been suggested for CuGeO$_3$ we find not only a worse description of the susceptibility but simultaneously evidence for discrepancies to the specific heat data.
REFERENCES

[1] J. Riera, and A. Dobry, Phys. Rev. B 51, 16098 (1995)
[2] G. Castilla, S. Chakravarty, and V. J. Emery, Phys. Rev. Lett. 75, 1823 (1995)
[3] B. Büchner, U. Ammerahl, T. Lorenz, W. Brenig, G. Dhalenne, A. Revcolevschi, Phys. Rev. Lett. 77, 1624 (1996)
[4] M. Hase, I. Terasaki, and K. Uchinokura, Phys. Rev. Lett. 70, 3651 (1993)
[5] I.S. Jacobs, J.W. Bray, H.R. Hart, Jr., L.V. Interrante, J.S. Kasper, and G.D. Watkins, Phys. Rev. B 14, 3036 (1976)
[6] L. N. Bulaevskii, A. I. Buzdin, and D. I. Khomskii, Solid State Commun. 27, 5 (1978)
[7] M.C. Cross, and D.S. Fisher, Phys. Rev. B 19, 402 (1979)
[8] E. Pytte, Phys. Rev. B 10, 4637 (1974)
[9] K. Hirota, D. E. Cox, J. E. Lorenzo, G. Shirane, J. M. Tranquada, M. Hase, K. Uchinokura, H. Kojima, Y. Shibuya, and I. Tanaka, Phys. Rev. Lett. 73, 736 (1994)
[10] M. Nishi, O. Fujita, and J. Akimitsu, Phys. Rev. B 50, 6508 (1994)
[11] C. K. Majumdar, and D. K. Ghosh, J. Math. Phys. 10, 1388 (1969)
[12] K. Okamoto, and T. Nomura, Phys. Lett. A 169, 433 (1992)
[13] J.C. Bonner, and M.E. Fisher, Phys. Rev. 135, A640 (1964)
[14] W. Geertsma, and D. I. Khomskii, Phys. Rev. B 54, 3011 (1996)
[15] J.P. Pouget, L.P. Regnault, M. Ain, B. Hennion, J.P. Renard, P. Veillet, G. Dhalenne, and A. Revcolevschi, Phys. Rev. Lett. 73, 736 (1994)
[16] K. Fabricius, A. Klümper, to be published
[17] M. Honda, T. Shibata, K. Kindo, S. Sugai, T. Takeuchi, and H. Hori, J. Phys. Soc. Jap. 65, 691 (1996)
[18] We mention that the uniaxial pressure dependences $\frac{\partial \chi}{\partial p_i}$ shown in Fig.2 of [3] and Fig.10 of [19] are given in wrong units ($10^{-7}$emu/gPa). The correct unit is $10^{-6}$emu/cm$^3$GPa.
[19] U. Ammerahl, T. Lorenz, B. Büchner, A. Revcolevschi, and G. Dhalenne, Phys. Rev. Lett. 78, 487 (1997)
[20] P. H. M. Loosdrecht, J. Zeman, G. Martinez, G. Dhalenne, and A. Revcolevschi, preprint
[21] B. Büchner, T. Lorenz (unpublished)
[22] X. Liu, J. Wosnitza, H.v. Löhneysen, and R.K. Kremer, Z. Phys. B 98, 163 (1995)
[23] M. Weiden, J. Köhler, G. Sparn, M. Köppen, M. Lang, C. Geibel, and F. Steglich, Z. Phys. B 98, 167 (1995)
[24] G. Uhrig, Preprint (Cologne 1997)
[25] H. Kuroe, J-I Sasaki, T. Sekine, N. Koide, Y. Sasago, K. Uchinokura, and M. Hase, Phys. Rev. B (in press)
[26] C.Gros, W. Wenzel, A. Fledderjohann, P. Lemmens, M. Fisher, G. Güntherodt, M. Weiden, C. Geibel, and F. Steglich, Magnon-magnon interaction in the Spin-Peierls compound CuGeO$_3$, Phys. Rev. B in press
[27] T. Lorenz, U. Ammerahl, R. Ziemas, B. Büchner, A. Revcolevschi, and G. Dhalenne, Phys. Rev. B 54, 15610 (1997)
[28] A. Klümper, Z. Phys. B 91, 507 (1993)
FIGURES

FIG. 1. Plot of numerical results for the susceptibility per site (in units of $J_1$) versus the reduced temperature $T/J_1$ for $\alpha = 0.1, \ldots, 0.4$, and analytical results for $\alpha = 0$ following [28] down to zero temperature. Shown in insets: behaviour of the susceptibility in the neighbourhood of the maximal values $\chi_{\text{max}}$ and plot of the corresponding temperature $T_{\text{max}}$ as a function of $\alpha$.

FIG. 2. Plot of numerical and analytical results for the specific heat per site similar to Fig.1.

FIG. 3. Plot of the maximal value of the magnetic susceptibility $\chi_{\text{max}}$ as a function of $\alpha = J_2/J_1$ (solid line) in the two parameter model. The experimental value (dashed line) is crossed at $\alpha = 0.354$. (The corresponding value of $T_{\text{max}}$ is 56K.)

FIG. 4. Depiction of the experimental results for the magnetic susceptibility in dependence on temperature (solid line). Also shown are the theoretical results for $\alpha = 0.354$ (dotted line) and $\alpha = 0.24$ (dashed line).

FIG. 5. Left upper panel: Experimentally observed specific heat of CuGeO$_3$ ($\circ$). The extrapolated low temperature phonon background $C_{\text{ph}} = \beta T^3$ with $\beta = 0.32mJ/K^4\text{mole}$ is indicated by the dashed line. The solid line shows the calculated magnetic specific heat for the exchange constants revealing the best fit to the magnetic susceptibility. Left lower panel: Experimentally observed specific heat of CuGeO$_3$ ($\circ$) and the estimated minimum magnetic specific heat (solid line, see text).
Right panel: Comparison between the minimum magnetic entropy as revealed from the specific heat data (●) and calculations of $S_{\text{mag}}$ assuming different exchange constants given in the figure. The dashed lines correspond to results of exact thermodynamic calculations for $\alpha = 0$ and the solid lines are obtained from numerical diagonalizations.
\[C(J/K\text{mole})\]

\[\beta T^3\]

\[J=80K, \alpha=0.35\]

\[S_{\text{mag}}(J/K\text{mole})\]

\[J=125K, \alpha=0.35\]

\[J=60K, \alpha=0\]

\[J=44K, \alpha=0\]

\[min. C_{\text{mag}}\]

\[min. S_{\text{mag}}\]
\[ \alpha = 0, 0.1, \ldots, 0.4 \]