Determination of Hamiltonian parameters of anisotropic one-dimensional quantum chains by susceptibility measurements

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With the use of finite-chain calculations, the perpendicular and parallel susceptibilities of anisotropic *XXZ* chains have been determined in all parameter domains of the model. It is shown how these results can be used to extract the parameters J, Δ, and g of the model unambiguously. Application to published materials validates the approach. If one considers all three components of the susceptibility ($\chi^{\parallel}$, $\chi^{\perp 1}$, $\chi^{\perp 2}$), they show a characteristic behavior in the different parameter regimes (ferromagnets and antiferromagnets, displaying planar- and uniaxial-anisotropic spin coupling, respectively). From the ratio $T^{\parallel}_{\max}/T^{\perp}_{\max}$ one can decide whether or not a given material is *XXZ*-like.

I. INTRODUCTION

Recently there has been renewed discussion of models of one-dimensional magnetic materials, both for quantum mechanical (e.g., eigenstates of the Hamiltonian) and thermodynamic properties (e.g., susceptibility, correlation functions) of the models. This is caused by the availability of more materials with one-dimensional magnetic character as well as by further advanced theoretical methods for the treatment of lattice theories. For a model described by the *XXZ* Hamiltonian for spin $\frac{1}{2}$,

$$H=2J\sum_{i=1}^{N}(S_i^xS_{i+1}^x+S_i^yS_{i+1}^y+\Delta S_i^zS_{i+1}^z)+\sum_{i=1}^{N}\vec{h}\cdot\vec{S}_i,\quad \vec{S}=\tfrac{1}{2}\vec{\sigma},\quad J>0 \tag{1}$$

we investigated the variation of static magnetic properties, such as the susceptibility parallel and perpendicular to the chain, with the anisotropy parameter Δ for all parameter domains. Results can be made plausible by the aid of a semiclassical interpretation of spin correlation. For the antiferromagnetic domain, a new method is proposed to determine the anisotropy parameter from experimental data. Former ambiguities are removed, and the procedure is not sensitive to the value of spectroscopic-splitting factors (g factors) and overall corrections (background) to the data. The ferromagnetic domain has also been studied and information on the behavior of the susceptibility and the determination of Δ is given. Fisher's exact results[1] on the antiferromagnetic Ising chain could be reproduced, as well as Takahashi's results[2] on the parallel susceptibility of planar chains which he obtained from the solution of a nonlinear integral equation derived by the Bethe ansatz. But his solution is possible only for some selected values of Δ.

Using experimental data published in the literature,[3–5] materials such as $CsCuCl_3$, $Cu(NH_3)_4SO_4\cdot H_2O$, $KCuF_3$,

$[C_6H_{11}NH_3]CuCl_3$ [=CHAC],

$[(CH_3)_3NH]XCl_3\cdot 2H_2O$, X=Co,Cu,

can be described by the (anisotropic) quantum *XXZ* chain. For CHAC and $[(CH_3)_3NH]CoCl_3\cdot 2H_2O$ interpretation as a slightly planar rather than Ising-like ferromagnet is proposed, contrary to.[5,6] $CsCoCl_3$ which has been regarded as an Ising-like antiferromagnet with $1/\Delta\approx 0.1$, does not seem to be an *XXZ*-like magnetic material at all. This might explain contradicting interpretations of elementary (dynamic) excitations in $CsCoCl_3$ which were published recently.[7]

All results were derived by finite-chain calculations. Extensive information is given in Ref. 8. Accuracy is believed to be high as has also been shown by previous investigations. The finite-chain calculations are discussed in Sec. II and the Appendix, results on susceptibilities are presented in Sec. III, and in Sec. IV we propose a new method to compare experimental and theoretical results and to obtain various parameters of the model. Our conclusions are presented in Sec. V.

II. FINITE-CHAIN CALCULATIONS

A. Spectrum

Finite-chain calculations have been used to find eigenstates and eigenvalues of the *XXZ* chain. This

approach has been pioneered by Bonner and Fisher[9] in the early sixties, and recently been used to determine neutron-cross sections by some groups.[10–12] By means of a computer, one numerically diagonalizes the Hamiltonian (1) for a finite number of lattice sites N. As there are 2^N basis states, one carefully has to exploit symmetries of the Hamiltonian.

(i) Translational invariance and invariance under spin rotations around the z axis render the momentum q and the total z component of spin

$$S_t^z=\sum_{i=1}^{N} S_i^z$$

as good quantum numbers. H, therefore, is of block diagonal form, and the size of the largest matrix to be diagonalized thus reduces from $2^N\times 2^N$ (e.g., $1024\times 1024\approx 10^6$ in the case of $N=10$) to approximately

$$\frac{1}{N}\begin{bmatrix} N \\ N_2 \end{bmatrix}\times\frac{1}{N}\begin{bmatrix} N \\ N_2 \end{bmatrix}$$

(i.e., 26×26 for $N=10$). This task can easily be fulfilled with present-day computers.

(ii) Owing to inversion symmetry only states with $0\le S_t^z\le N/2$, due to reflection symmetry, only states with momentum $0\le q\le\pi$ have to be calculated. Therefore, a large part of (S_t^z,q) blocks of H need not be diagonalized and explicitly determined. Eigenstates of negative S_t^z are obtained from the respective positive S_t^z states by inversion of the spins (except for phases).

Eigenstates of momentum q larger π are obtained from the respective states of momentum $2\pi-q$ by complex conjugation of their coefficients (in a basis of translational invariant Ising-basis states, e.g., $|\alpha,q\rangle$, to obtain the eigenvector with momentum $q'=2\pi-q$, all $|\alpha,q\rangle$ are replaced by $|\alpha,2\pi-q\rangle$ and the new coefficients are the complex conjugates of the old ones).

These symmetries and invariances help to serve a good amount of computer time and storage. Unfortunately for numeric calculations, H is given in operator form, so in a first step the matrix H has to be calculated in a suitable representation. Great reductions of computer time in this very time consuming process [$\frac{1}{2}(2^N)^2 6N$ spin operators have to be applied] can be made by application of Lanczos's method[13] (see the Appendix), which for a $m\times m$ matrix directly leads to a tridiagonal form in $m-1$ operations only (compared to $m\times m$ operations plus explicit tridiagonalization).

B. Susceptibility

Having calculated all eigenvalues and eigenstates, expressions for the physical susceptibility were obtained from

$$\chi^i=\lim_{h_i\to 0}\frac{\partial^2 F(T,h)}{\partial h_i^2},\quad i=x,y,z \text{ or } ||,\perp \tag{2}$$

where $F(T,h)$ is the free energy of the chain. Owing to the quantum mechanical structure of the model, the susceptibility is not isotropic (except at $\Delta=\pm 1$), and one discriminates the susceptibility $\chi^{||}=\chi^z$ parallel to the chain, and a perpendicular susceptibility $\chi^{\perp}=\chi^x=\chi^y$, if a field perpendicular to the chain is applied. Direct evaluation of (2) gives

$$\chi^{||}(T)=N_L(g_{||}\mu_B)^2\beta\int d\omega\, S^{||}(0,\omega) \tag{3}$$

while perturbation calculations[14] and Hohenberg and Brinkman's exact sum rules[15] both give

$$\chi^{\perp}(T)=N_L(g_{\perp}\mu_B)^2\frac{1}{|J|}2\int\frac{d\omega}{\omega}S^{\perp}(0,\omega)\ . \tag{4}$$

N_L is Avogadro's number, μ_B the Bohr magneton, $\beta=1/kT$, g the spectroscopic splitting factor, and $S^v(q,\omega)$ the dynamic form factor

$$S^v(q,\omega)=\frac{1}{2\pi N}\sum_{l,m}e^{iqm}\int dt\, e^{i\omega t}\langle S_l^v(t)S_{l+m}^v\,|\,(0),\rangle,$$

$$v=\begin{cases}||\,\hat{=}\,z\\ \perp\,\hat{=}\,x,y\end{cases} \tag{5}$$

N equals the chain length. Here the angular brackets denote the thermodynamic average,

$$\langle AB\rangle=\frac{1}{Z}\mathrm{tr}(e^{-\beta H}AB)\ . \tag{6}$$

By using a set of eigenstates, $\{|\lambda\rangle,\lambda=1,\ldots,2^N\}$ with eigenvalues E_λ, we obtain

$$S^v(q,\omega)=\frac{1}{Z}\sum_{\lambda,\lambda'}e^{-\beta E_\lambda}\delta(\omega-(E_{\lambda'}-E_\lambda))\times|\langle\lambda|S^v(q)|\lambda'\rangle|^2\ , \tag{7}$$

where

$$S^v(q)=\frac{1}{\sqrt{N}}\sum_l e^{-iql}S_l^v\ .$$

Matrix elements of the type $\langle\lambda|S^z(0)|\lambda'\rangle$ have to be calculated to find $\chi^{||}$ and matrix elements of type $\langle\lambda|S^+(0)|\lambda'\rangle$ are necessary for calculation of $\chi^{\perp}$, where one takes advantage of the relations

$$S^x=\tfrac{1}{2}(S^+ + S^-)\ , \tag{8a}$$

$$\langle\lambda|S^-(q)|\lambda'\rangle=\langle\lambda'|S^+(-q)|\lambda\rangle^*\ . \tag{8b}$$

Selection rules can be used to greatly reduce the

number of matrix elements to be calculated:

$$\langle\lambda\,|\,S^{+}(q)\,|\,\lambda'\rangle\sim\delta_{S^z_\lambda,S^z_{\lambda'}}\ , \tag{9a}$$

$$\langle\lambda\,|\,S^{+}(q)\,|\,\lambda'\rangle\sim\delta_{S^z_\lambda,S^z_{\lambda'+1}}\ , \tag{9b}$$

$$\langle\lambda\,|\,S^{\pm,z}(0)\,|\,\lambda'\rangle\sim\delta_{q_\lambda,q_{\lambda'}}\ . \tag{9c}$$

Owing to the inversion and reflection symmetry, one again does not have to calculate all matrix elements left by the rules [9(a)—9(c)]. Only a minor part of the $2^N\times 2^N$ matrix elements actually has to be determined. For chains of the length 8 and 10 no further "computing tricks" had to be applied, and calculation times kept in the range of seconds (chain length 8, Univac 1100 machine).

C. Accuracy

Several authors, including Bonner and Fisher, investigated how finite-chain results extrapolated to $N=\infty$.[9,16] Concerning static properties (groundstate energies, integrated amplitudes, autocorrelation functions, susceptibilities) one obtains results surprisingly close to the $N=\infty$ limit even at very low N, and good convergence of results (see extensive treatment by Blöte, Bonner, and Fisher).

For the few exact results, finite-chain calculations at $N=10$ differ by only 0.3% from the exact values for infinite chains. The results presented in the following thus are either extrapolated or corrected by an overall factor. Therefore we believe our results to be of high accuracy in representing the infinite chain, the actual computational error being negligible.

III. SUSCEPTIBILITY RESULTS

Directions in which spin coupling is dominant (Ising-like, ||; planar, $\perp$) will be denoted as "hard" directions as opposed to "soft" interactions (Ising-like, $\perp$; planar, ||). The exchange parameter J will be taken to be positive throughout the following sections. Ferromagnetic chains then correspond to $\Delta<0$, antiferromagnetic chains to $\Delta>0$ due to the operator U which transforms $-H(\Delta)$ into[17]

$$H(-\Delta)=-UH(\Delta)U^{-1}\ .$$

Figure 1(a)—1(g) show the parallel and perpendicular susceptibilities for some selected values of Δ in the parameter domains of ferromagnets (antiferromagnets) [$\Delta<0$ ($\Delta>0$)] and planar (Ising-like) chains [$|\Delta|<1$ ($|\Delta|>1$)]. The dimensionless quantities

$$\chi^r=\chi J/N_L g^2\mu_B^2 \tag{10a}$$

are shown against the relative temperature

$$T^r=kT/J\ . \tag{10b}$$

For the planar ferromagnet and the antiferromagnet [Fig. 1(a)] the parallel susceptibility displays high and narrow maxima at low temperature. As Δ increases from -1 (isotropic ferromagnet), these maximum move towards higher temperatures, decreases in height and are broadened. The perpendicular susceptibility of the antiferromagnet [Fig. 1(b)] shows the same behavior but the change in shape of the curves is less pronounced. For both types of susceptibilities (||,$\perp$) the maxima move towards infinity as Δ increases, but at a different rate. This observation will be the basis for the new method to determine J,Δ,g proposed in Sec. IV. The parametrization of the model,

$$H=2J'\sum\left[\frac{1}{\Delta}(S_i^xS_{i+1}^x+S_i^yS_{i+1}^y)+S_i^zS_{i+1}^z\right]$$

is helpful in the Ising-limit, where one avoids the very broad maxima shown in Figs. 1(a) and 1(d) thus making the determination of $\chi^r_{\max}$ and $T^r_{\max}$ easier and more accurate. Nevertheless all data are given in terms of Hamiltonian (1).

As can be expected from a pure Ising ferromagnet (which has a ferromagnetic groundstate as opposed to the planar ferromagnet which has an antiferromagnet ground state) $\chi^{||}$ of the Ising-like ferromagnet diverges as $T\to 0$ [Fig. 1(c)]. For $\chi^{\perp}$ of the planar ferromagnet we find a divergent behavior too, Fig. 1(c).

In the soft direction of the Ising-like ferromagnet, $\chi^{\perp}$ shows maxima [Figs. 1(d) and 1(g)] which similarly to $\chi^{||}$ of the planar ferromagnet increase in height and move towards $T=0$ as Δ increases from $-\infty$, till at $\Delta=-1$ we find a power-law divergence (being the isotropic model, $\chi^{\perp}$ has to display the same behavior as $\chi^{||}$ at this value). For the hard directions of the ferromagnet (Ising-like, ||; planar, $\perp$) the divergence is nearly independent of Δ [Figs. 1(e) and 1(f)], as is shown in a log-log plot. This is not too surprising because anyway maximum spin coupling will be found in these directions so that alignment should not be too sensitive to the actual strength of the strong (hard) coupling. In both cases as $T\to 0$ we find a power-law divergence. Compared to the curves calculated by Takahashi[2] for $\chi^{||}$, using nonlinear integral equations derived from the Bethe ansatz, and to the maxima of $\chi^{||}$ and $\chi^{\perp}$ of the antiferromagnetic Ising chain calculated by Fisher,[1] we find small deviations for temperatures $kT/J<0.2$ which are due to finite-chain effects (see above). Position and height of maxima close to $\Delta=-1$ therefore show only the general trend. At

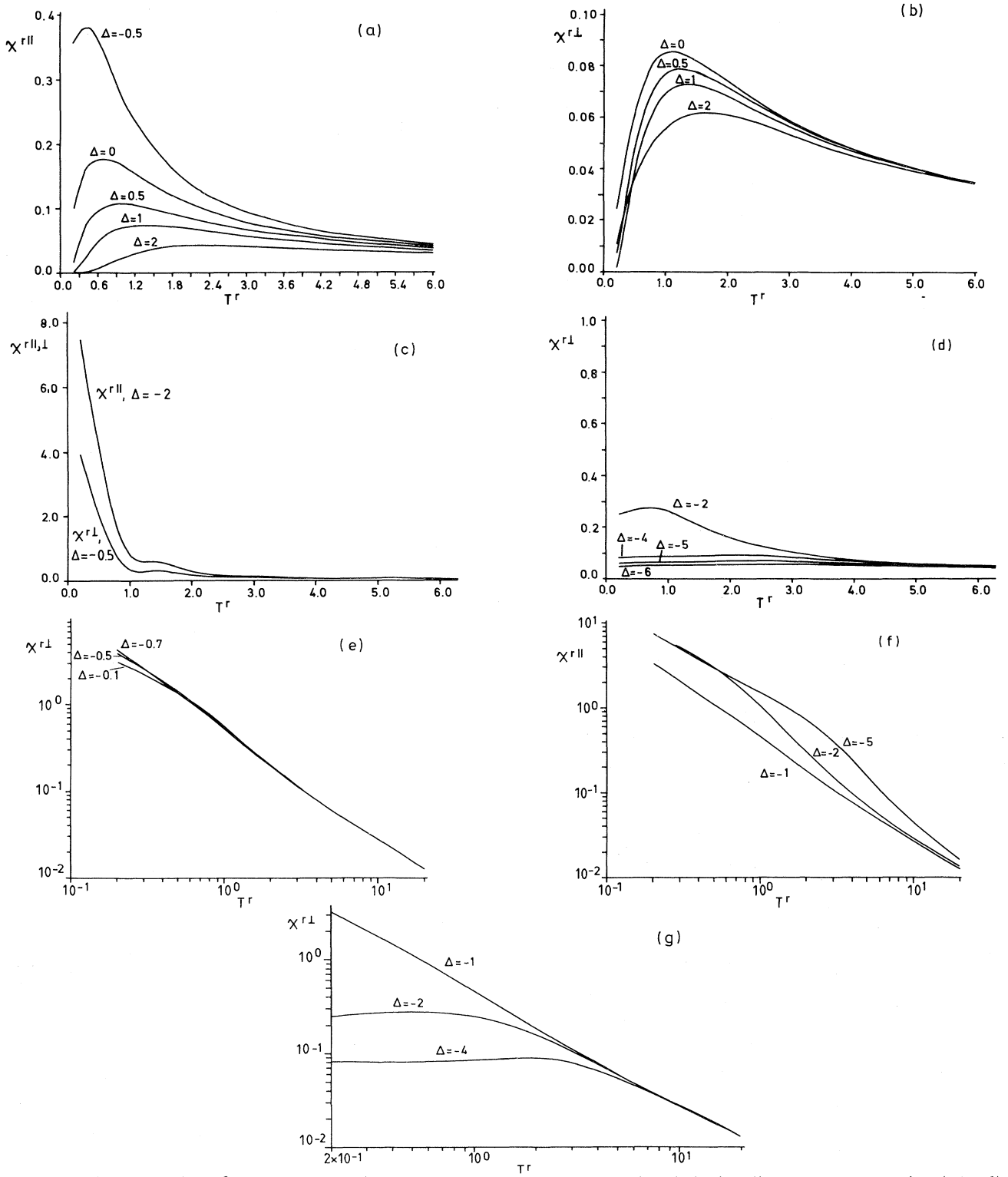


FIG. 1. Susceptibility of the XXZ model parallel and perpendicular to the chain in all parameter domains ($N=6$) versus $T^r(=kT/J)$: (a) parallel susceptibility of planar ferromagnet, planar, and Ising-like antiferromagnet, (b) perpendicular susceptibility of the antiferromagnet, (c) perpendicular susceptibility of the planar ferromagnet and parallel susceptibility of the Ising-like ferromagnet, (d) perpendicular susceptibility of Ising-like ferromagnets, (e) perpendicular susceptibility of planar ferromagnets, (f) parallel susceptibility of Ising-like ferromagnets, and (g) perpendicular susceptibility of the Ising-like ferromagnet with $\Delta \leq -1$.

$\Delta=-1$ the behavior of $\chi^{\|}$ changes from a very high and narrow (antiferromagnetic) peak close to $T=0$ to the diverging type which is expected for a ferromagnet, similar as $\chi^{\perp}$ when moving from $\Delta=-\infty$ to -1 [Fig. 1(g)].

From a physical point of view these results can easily be made plausible by considerations of spin interaction. The Ising-like ferromagnet shows long-range order at $T=0$, the susceptibility therefore diverges. Nevertheless average spin projections into perpendicular directions can be zero, $\chi^{\perp}$ of the Ising-like ferromagnet therefore displays maxima. The antiferromagnet, displaying staggered long-range order shows no spontaneous magnetization at $T=0$, therefore we expect maxima at finite T. Now consider the planar case: Both ferromagnet and antiferromagnet display an antiferromagnetic ground state in this domain, $S_t^z=0$. For the parallel susceptibility we therefore find the same qualitative behavior as in the antiferromagnetic regime. As for the perpendicular susceptibility, even under the condition $S_t^z=0$ the spins can order ferromagnetically or antiferromagnetically in the xy plane for the planar model. So this argument supports the existence of a divergence (maximum) in the planar ferromagnetic (antiferromagnetic) regime for $\chi^{\perp}$.

IV. DETERMINATION OF EXPERIMENTAL PARAMETERS BY THEORY

In the following, $T_{\max}$ always denotes the position of the maximum, and $\chi_{\max}$ denotes the value of the susceptibility at the maximum. Quantities without index r denote values measured in experiment, while the index r denotes finite-chain results [cf. Eqs. 10(a) and 10(b)]. Up to now, information on maximum positions existed only for a few isolated values of Δ and experimental data were either fitted to the isotropic model (Bonner and Fisher's results), to the Ising model (using Fisher's calculations), or to the XY chain. But these determinations still contain a lot of ambiguity, as Fig. 2 shows. The same experimental points can both be fitted with J high, $\Delta=0$, or with J small, and $\Delta=0.25$.

Though distinct, both curves lie very close together and coincide within experimental error. No suggestion can be made whether deviations from the theoretical curves are caused by anisotropy, bad one-dimensional character, crystal fields, etc. This behavior is especially pronounced in the hard directions.

From extensive numerical studies we therefore propose the following unambiguous method to determine Δ and J from susceptibility measurements, starting from the knowledge of the position

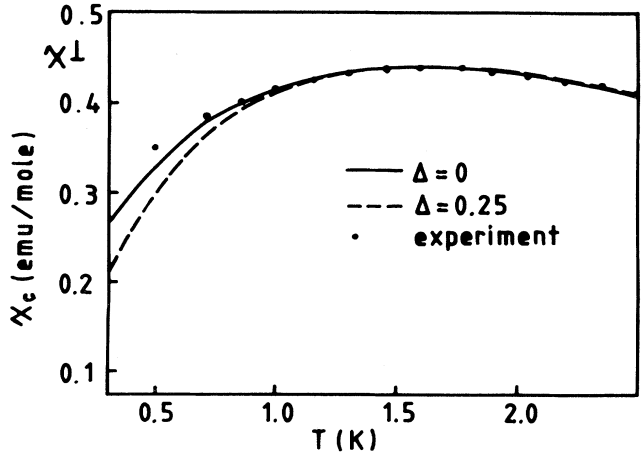


FIG. 2. Perpendicular susceptibility of $CsCoCl_4$ (Ref. 14). The planar antiferromagnet chain with $\Delta=0$ and $\Delta=0.25$ has been fitted (hard directions, therefore only small deviations among the curves).

of the maxima in both $\|$ and $\perp$ direction (i.e., $T^{\|}_{\max}/T^{\perp}_{\max}$). It is applicable in the antiferromagnetic regime only but does not require the knowledge of the g factors which rather can be determined this way.

Figures 3(a)–3(c) show the ratio $T^{\|}_{\max}/T^{\perp}_{\max}=T^{r\|}_{\max}/T^{r\perp}_{\max}$ for the planar or Ising-like antiferromagnet, as determined by finite-chain calculations. This ratio is a function of Δ only. Experimental knowledge of the position of maxima of $\chi^{\|},\chi^{\perp}$ therefore suffices to determine Δ. The procedure is not sensitive to background or overall corrections. J then results from the position of the $\chi^{\|}$ or $\chi^{\perp}$ maximum, as given by $J=kT_{\max}/T^{r}_{\max}(\Delta)$. The more accurate measurement ($\|,\perp$) can be used to determine J. Values of $T^{r\|,\perp}_{\max}$ vs Δ are given in Figs. 3(d)–3(g) for the measurement in a parallel resp. perpendicular direction. The absolute heights of $\chi^{r}_{\max}T^{r}_{\max}$ (Figs. 4 and 5) serve to calculate the g factors appropriate to the material concerned [Eq. (11)]. Results obtained this way can then be compared with specific heat or other measurements for the same material.

We are able to determine J and Δ independent of g. This is of special advantage because J and Δ are the characterizing parameters of the XXZ model which are important for comparisons with nonmagnetic properties (specific heat, internal energy, etc.). The g factor is the only quantity influenced by temperature-independent correction or background, e.g., the absolute height of the susceptibility χ.

In the case that separate measurements of $\chi^{\perp},\chi^{\|}$ are not possible, e.g., because of unknown crystal structure or unknown mixture of $\chi^{\|}$ and $\chi^{\perp}$ in the

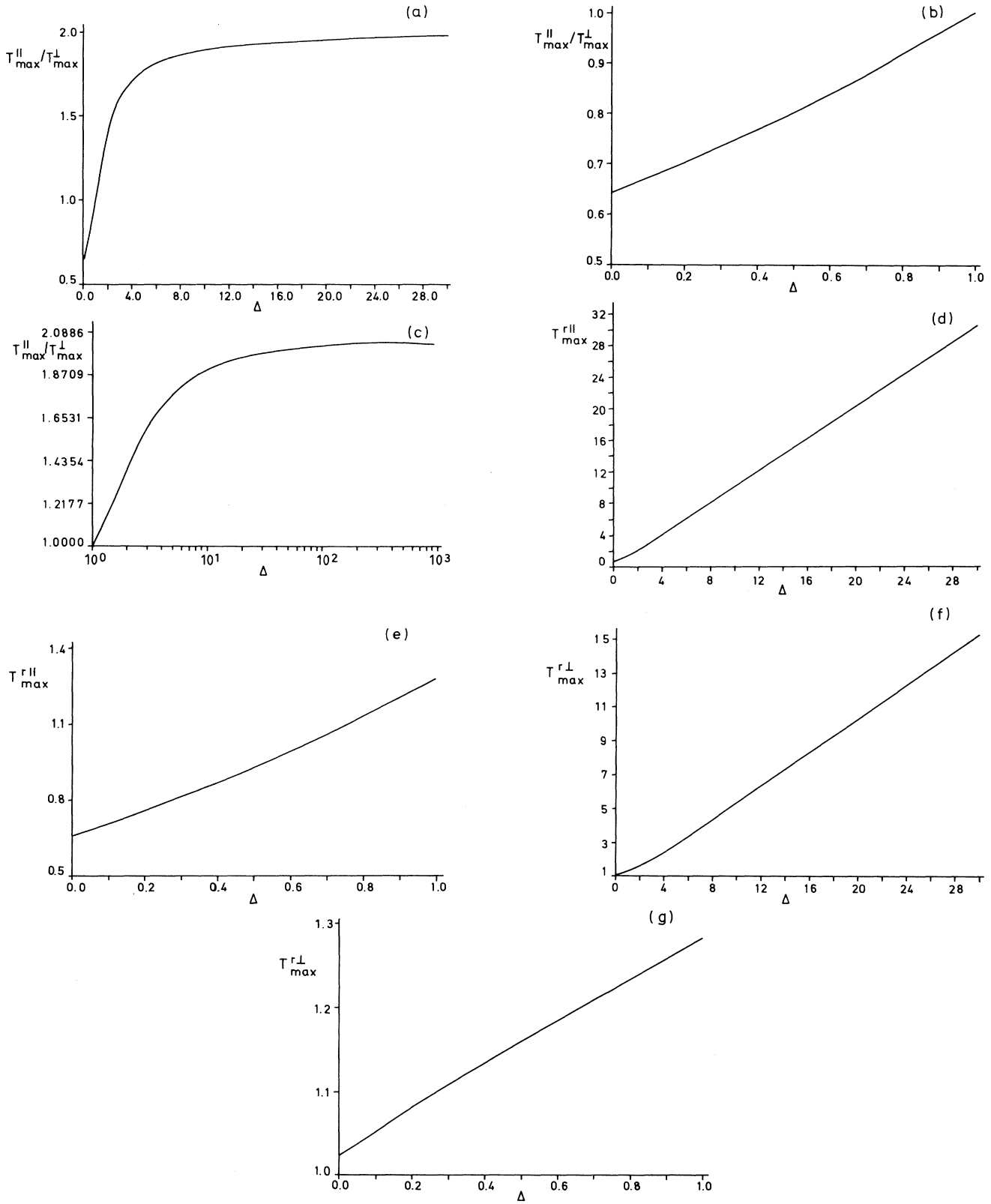


FIG. 3. (a) Ratio $T^{\parallel}_{max}/T^{\perp}_{max}$ of the antiferromagnetic XXZ chain, $0 \leq \Delta \leq 30$; (b) as (a), for the planar domain; (c) as (b), for the Ising-like domain; (d) position of maximum of $\chi^{r\parallel}$; (e) as (d), for the planar domain; (f) position of maximum of $\chi^{r\perp}$; (g) as (f), for the planar domain.

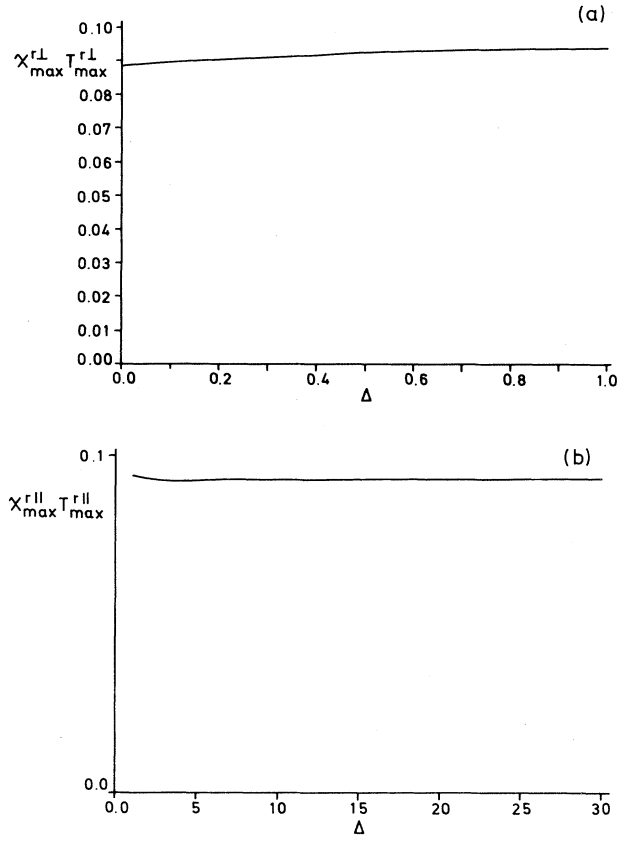


FIG. 4. $\chi^r_{max}\, T^r_{max}$ as determined for hard directions. (a) Planar, $\chi^{r\perp}_{max} T^{r}_{\perp max}$; (b) Ising-like, $\chi^{r||}_{max} T^{r||}_{max}$.

other crystal directions one has to determine J and Δ from the knowledge of the absolute height of the susceptibility measured and the position of the maximum (see below). But this procedure is strongly sensitive to the accuracy of the g factors and to background corrections of χ. Both J and Δ can be affected by errors in g.

Nevertheless we can provide new numerical results which make the determination of intermediate values of Δ possible. The best way seems to us to be the following: Equation (10) could serve to calculate J if Δ were known already. Because this is not the case, we replace J in (10a) by (10b):

$$\chi^r = \chi \frac{kT}{T^r} \Big/ N_L g^2 \mu_B^2 \,. \tag{11}$$

Equation (11) now is applied at the position of the maximum:

$$\frac{\chi_{max} T_{max} k}{N_L g^2 \mu_B^2} = \chi^r_{max}(\Delta) T^r_{max}(\Delta) \,. \tag{12}$$

The right-hand side is a function of the anisotropy parameter chosen, the left-hand side is given by experiment. $\chi^r_{max} T^r_{max}$ vs Δ is shown in Figs. 4 and 5 for the parallel and perpendicular directions. $\chi^r_{max}(\Delta) T^r_{max}(\Delta)$ monotonically increases (decreases) for $\perp$ ($||$) with Δ. If $\chi_{max} T_{max}$ and g are known with reasonable accuracy one can therefore use their absolute value to determine Δ, choosing the appropriate figure J then follows from Eq. (10b), using Figs. 3(d)—3(g) and 6(a) and 6(b).

But as Figs. 2 and 4 show, data on hard directions cannot serve to extract both J and Δ: variation of curves is smaller than experimental error, and only one parameter can be calculated if the other is known already. This is not a drawback of our method but rather a special property of the XXZ model: its magnetic properties in hard directions are insensitive to the actual strength of the coupling. Nevertheless $\chi_{max} T_{max}$ says whether the measurement corresponds to a hard direction: one only cannot decide between different parameter pairs in this direction. This method of determination has to be applied if data on only one direction are known, or in the ferromagnetic regime where only the soft direction susceptibility displays maxima.

V. CONCLUSIONS

Our results can be summarized as follows (Fig. 7). The susceptibility of antiferromagnetic XXZ chains displays maxima, both measured parallel or perpendicular to the chain axis. The ratio of the temperatures at the maxima, $T^{||}_{max}/T^{\perp}_{max}$, cannot exceed 2.404 but is larger than 0.64. Ferromagnets display a more complicated behavior: In the hard directions (Ising-type, $||$; planar, $\perp$), the susceptibility always diverges as $T \to 0$. (All spins point into this direction, forced through the strong interaction.) In soft directions (when the averaged spin component is zero then), the ferromagnet shows antiferromagnetic features, i.e., the susceptibility stays finite and has a maximum. Only at $\Delta = -1$ both $\chi^{\perp}, \chi^{||}$ diverge.

Owing to anisotropic g factors the curves for the perpendicular directions can split into two (Fig. 7). The anisotropy of the spin interaction causes the occurrence of two different maxima resp. maximum and divergence of $\chi^{||}, \chi^{\perp}$. The anisotropy of the g factors causes the splitting of the perpendicular susceptibility.

We developed a new method to extract the Hamiltonian parameters from measurements which we applied to data published in the literature (see Table I, Fig. 8). As for $KCuF_3$ (Ref. 4), we can confirm the results on J and Δ: $KCuF_3$ is an isotropic antiferromagnet, with $J/k = 190$ K. Here $\chi^{||}$ is shifted relative to $\chi^{\perp}$ due to anisotropic g factors [see Fig. 8(a)]. Both $CsCuCl_3$ (Ref. 19) and

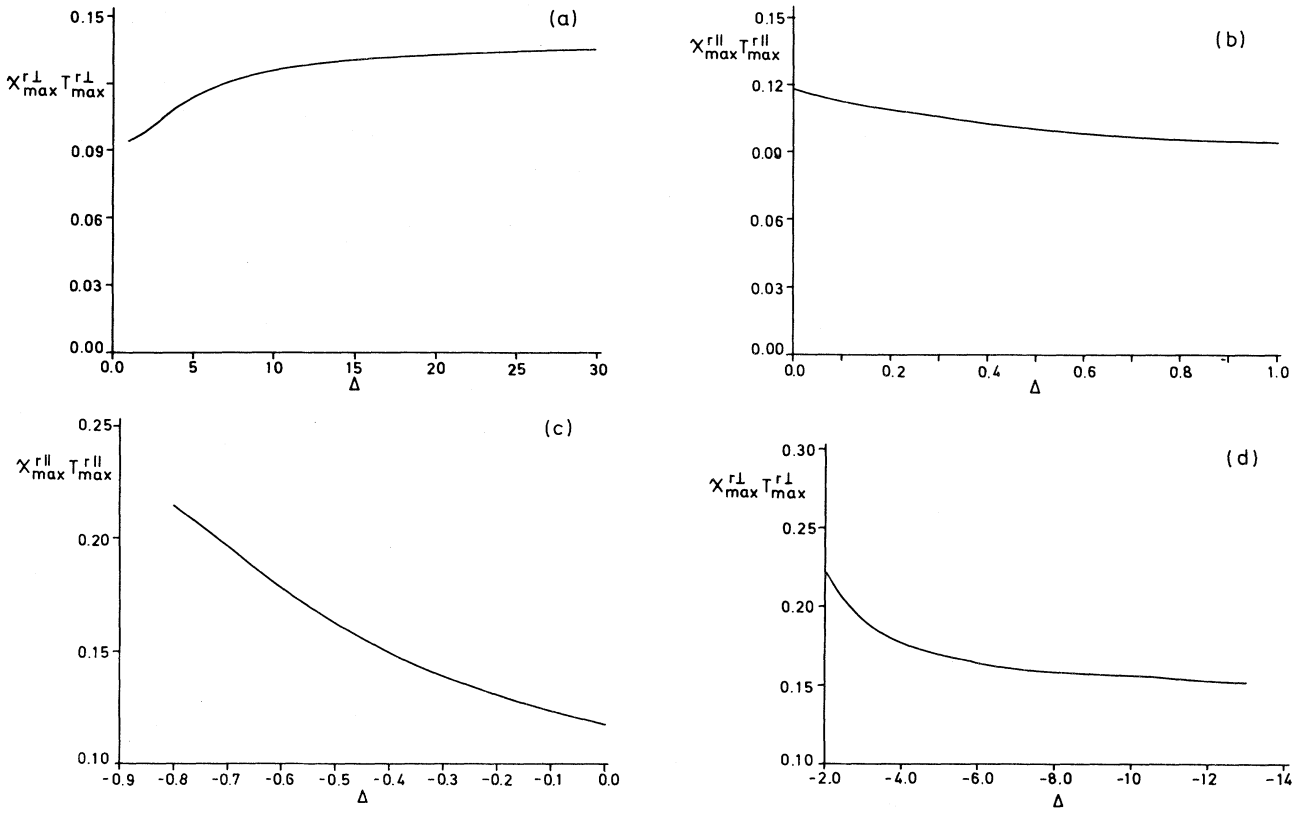


FIG. 5. $\chi^{r}_{\max}T^{r}_{\max}$, as determined for soft directions: (a) Ising-like, $\chi^{r\perp}_{\max}T^{r\perp}_{\max}$; (b) planar, $\chi^{r\parallel}_{\max}T^{r\parallel}_{\max}$; (c) planar ferromagnet, $\chi^{r\parallel}_{\max}T^{r\parallel}_{\max}$; (d) Ising-like ferromagnet, $\chi^{r\perp}_{\max}T^{r\perp}_{\max}$.

$Cu(NH_3)_4SO_4\cdot H_2O$ (Ref. 3) are slightly Ising-like antiferromagnets, with $\Delta=1.36$ and $\Delta=1.22$, respectively. Using these values we have also determined the parameters J, $g^{\parallel}$, and $g^{\perp}$. Now let us look at $CsCoCl_3$, which attracted the experimentalist's interest in order to discriminate between the theories of elementary excitation of des Cloiseaux and Gaudin[17] or Ishimura and Shiba[10] in Ising-type antiferromagnets. Here $\chi^{\parallel}$ has a maximum at 85 K, from $T^{\parallel}_{\max}/T^{\perp}_{\max}\leq 2.404$ we see that $T^{\perp}_{\max}$ should be $\gtrsim 35$ K, if $CsCoCl_3$ represents a one-dimensional $\widetilde{XXZ}$ model in this temperature range. We can find a maximum of $\chi^{\perp}$ at 17 K,[19] well below the temperature predicted by the XXZ model, and even below the transition temperature $T_N=25$ K to higher dimensional ordering.

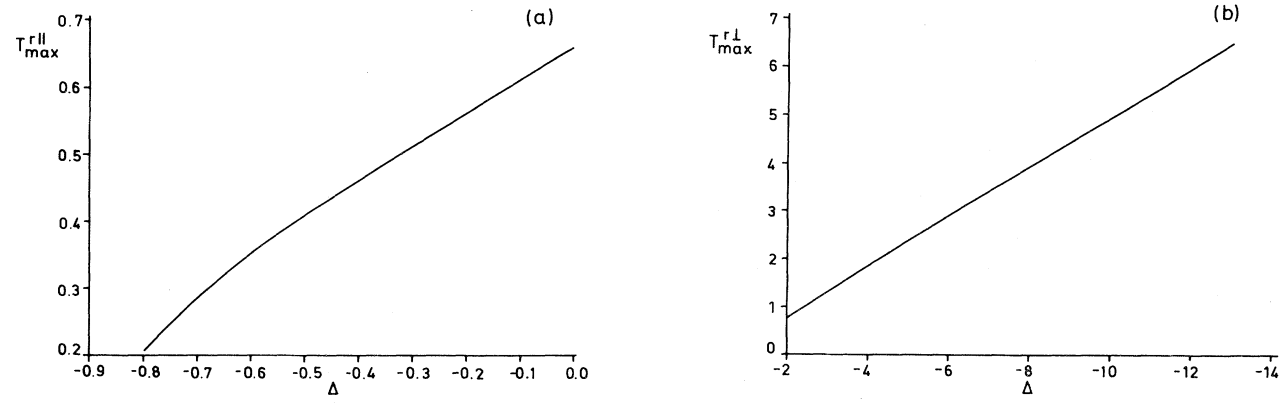


FIG. 6. Position of maximum temperature for soft direction of ferromagnets: (a) Planar ferromagnet, $T^{r\parallel}_{\max}$; (b) Ising-like ferromagnet, $T^{r\perp}_{\max}$.

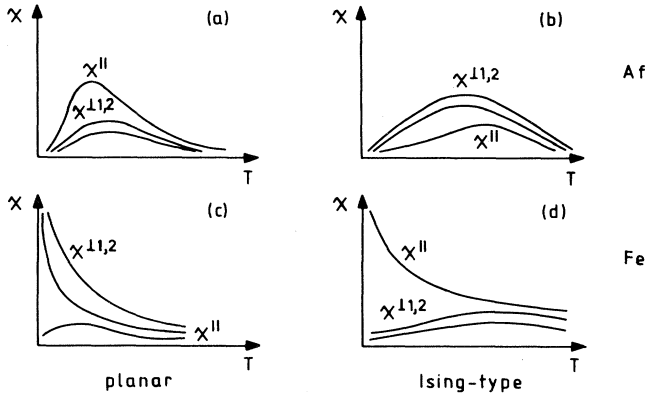


FIG. 7. Principal behavior of the susceptibilities: (a) Planar antiferromagnet, (b) Ising-like antiferromagnet, (c) planar ferromagnet, (d) Ising-like ferromagnet.

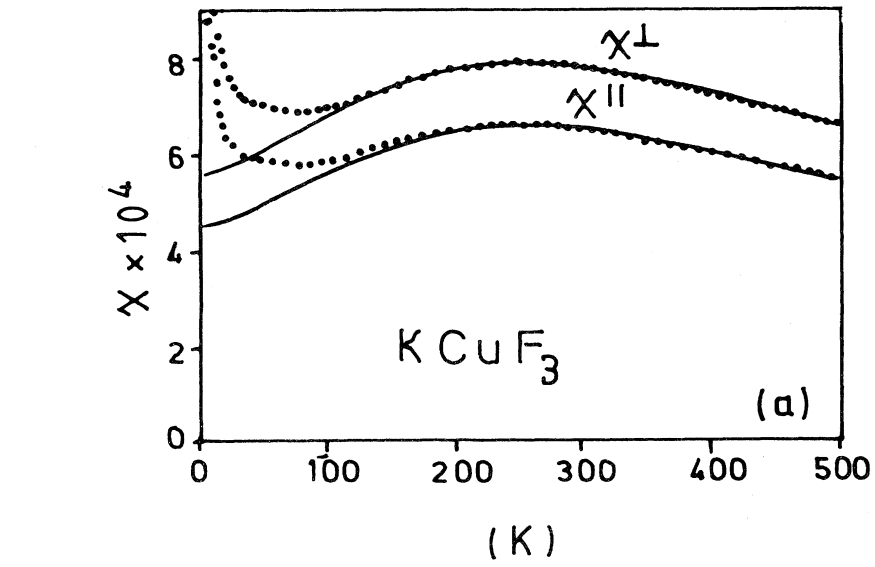
Thus we can either conclude that $CsCoCl_3$ behaves higher dimensional even in a temperature range extending beyond $T_N = 25$ K and also beyond $T^{\perp}_{max} \geq 35$ K, because this maximum is suppressed. Or we have to assume that $CsCoCl_3$ is one-dimensional above T_N, but then cannot be governed by *XXZ*-like behavior until well above $T = 35$ K.

$\chi^{||}$ of the ferromagnet $[(CH_3)_3NH]CuCl_3 \cdot 2H_2O$ could be fitted equally well to the Ising chain or the isotropic Heisenberg chain (see Ref. 18). But from the divergence of $\chi^{\perp}, \chi^{||}$, Fig. 8(b), we note that this material must be an isotropic ferromagnet. Here $\chi^{\perp_1}$ and $\chi^{\perp_2}$ are split probably due to anisotropic g factors. Next CHAC is slightly planar rather slightly Ising-like[5] as can be seen in Fig. 8(c). It displays a maximum measured parallel to the chain, while the perpendicular susceptibilities diverge (again split due to g anisotropy).

Although the identification of the soft direction of the ferromagnet is easy (it is the direction in which there is a maximum), one still has to distinguish between the planar and the Ising-like case, because the magnetic behavior in both domains is quite similar. If the soft direction is perpendicular, one observes two maxima—and therefore the system is Ising-like; if the soft direction is parallel, one observes only one maximum but two divergences and the system is planar (see Fig. 7). From this reasoning we suggest that $[(CH_3)_3NH]CoCl_3 \cdot 2H_2O$ is a planar ferromagnet. We determine $\Delta = -0.08$ and $J/k = 8.2$ K. Experimental data further show that $\chi^c/\chi^a = \chi^{\perp_1}/\chi^{\perp_2} \cong \text{const} = g^2_{\perp_2}/g^2_{\perp_1}$, consistent with the existence of a divergence which is split by anisotropic g factors. According to the duality of the planar and Ising-like model which will be discussed elsewhere one could equally fit $\chi^{\perp}$ of the Ising model

TABLE I. Comparison of published values of parameters with those determined in the present work.

					As published in literature				As determined here			
	$T^{||}_{max}$	$T^{\perp}_{max}$	$\chi^{||}_{max}$ (emu/mole)	$\chi^{\perp}_{max}$ (emu/mole)	J/k	Δ	$g^{||}$	$g^{\perp}$	J/k	Δ	$g^{||}$	$g^{\perp}$
(Ref. 4) $KCuF_3$	243 K	243 K	6.7×10^{-4}	8×10^{-4}	190 K	1	2.14	2.27	190 K	1	2.20	2.40
(Ref. 19) $CsCuCl_3$	9 K	7.7 K	18×10^{-3}	21×10^{-3}	7 K	1?	2.19	2.19	5.62 K	1.36	2.20	2.18
(Ref. 3) $Cu(NH_3)_4SO_4 \cdot H_2O$	3.63 K	3.23 K	4×10^{-2}	$3.89/3.61\times10^{-2}$	3.15 K	1	2.11	2.0/2.05	2.43 K	1.22	2.08	1.93/1.86
(Ref. 20) $[(CH_3)_3NH]CoCl_3 \cdot 2H_2O$	5 K		0.1128		7.7	$-\infty$	3.59	3.31/7.55	8.2 K	−0.08		



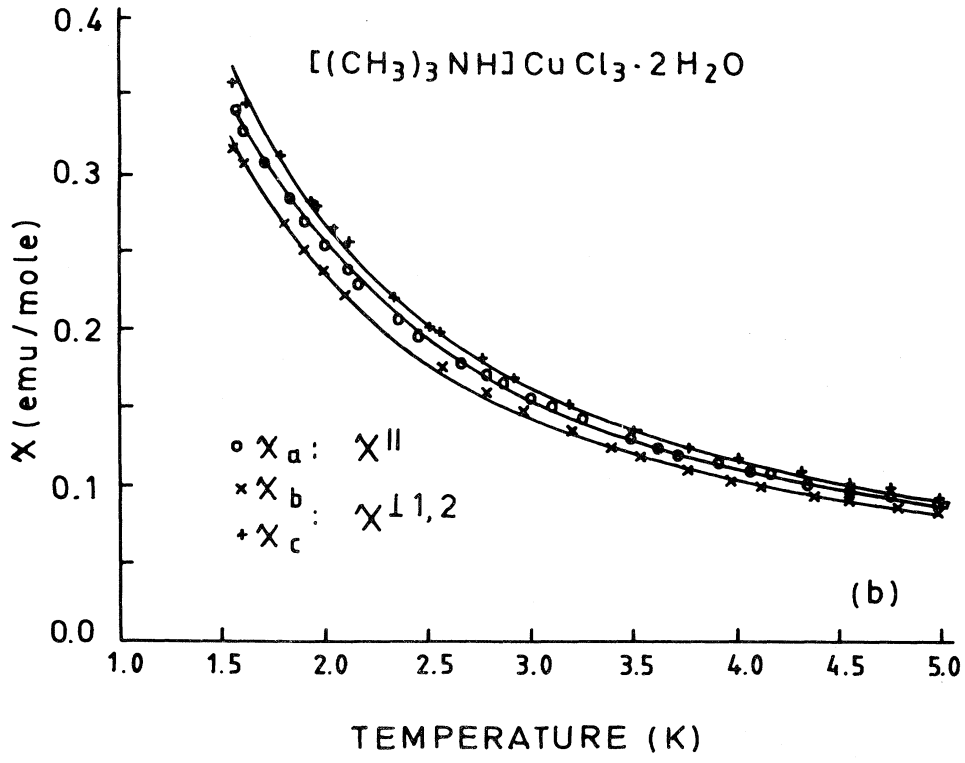


FIG. 8. (a) Heisenberg linear antiferromagnetic chain, fitted to $KCuF_3$, (b) data points of $[(CH_3)_3NH]CuCl_3 \cdot 2H_2O$, an isotropic ferromagnet, (c) data points of CHAC, displaying the planar character of this ferromagnet, (d) magnetic susceptibility of $[(CH_3)_3NH]CoCl_3 \cdot 2H_2O$, a planar ferromagnet.

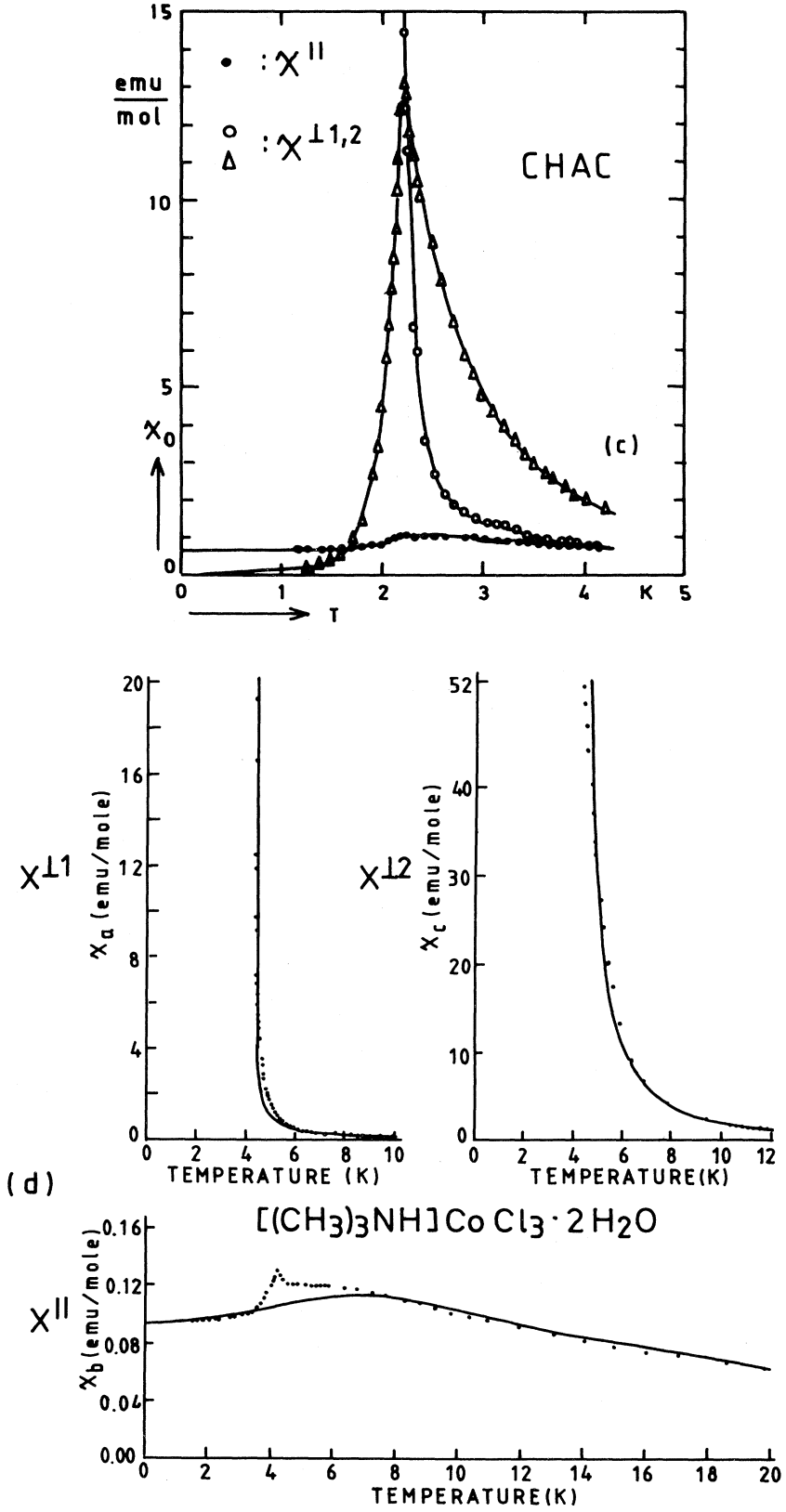


FIG. 8. (*Continued.*)

to $\chi^{||}$ measured as has been done by the authors in Ref. 6. Of course this would predict two divergences and does not correspond to the measurements.

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APPENDIX

Lanczos's method[13,20] for matrix diagonalization will shortly be described. As noted above, its major advantage is the fact that H can be given in operator form. The method consists in determining a special basis in which H has tridiagonal form. It is an iterational method which stops after $n-1$ steps (compared to n^2 applications of H, if one evaluates the full matrix). Let $|1\rangle$ be an arbitrary base vector, and apply H to $|1\rangle$:

$$H|1\rangle = a_1|1\rangle + b_1|2\rangle \,, \tag{A1a}$$

$$H|2\rangle = c_1|1\rangle + a_2|2\rangle + b_2|3\rangle \,. \tag{A1b}$$

Thus new base vectors $|2\rangle, |3\rangle$ are generated by orthogonal decomposition. Repeated applications of this process always render $|u\rangle = H|i\rangle$ as linear combinations of the base vectors already obtained, and a new base vector $|i+1\rangle$ orthogonal to all previous $|i\rangle$ as long as $\{|i\rangle\}$ is not complete. After the nth step one obtains

$$H|n\rangle = \sum_{i=1}^{n-2} \alpha_i |i\rangle + c_{n-1}|n-1\rangle + a_n|n\rangle - b_n|n+1\rangle \,. \tag{A2}$$

Some simple considerations show the α_i to be zero: The scalar product of $H|n\rangle$ with $\langle j|$, $j \le n-2$, yields

$$\langle j|H|n\rangle = \alpha_j = (\langle j|H)|n\rangle \,, \tag{A3}$$

H being Hermitian. But $H|j\rangle$ can only be a linear combination of states $1, \ldots, j+1$, therefore the scalar product has to be zero.

Our operations thus give the following structure:

$$\begin{aligned} H|1\rangle &= a_1|1\rangle + b_1|2\rangle \,, \\ H|n\rangle &= c_{n-1}|n-1\rangle + a_n|n\rangle + b_n|n+1\rangle \end{aligned} \tag{A4}$$

and the whole process terminates if $b_n \equiv 0$. The matrix H in this new base $|n\rangle$ is tridiagonal and $b_i = c_i$ because of Hermiticity:

$$H = \begin{bmatrix} a_1 & b_1 & 0 & 0 & \cdots \\ b_1 & a_2 & b_2 & 0 & \cdots \\ 0 & b_2 & a_3 & b_3 & \\ 0 & 0 & b_3 & \cdots & \end{bmatrix} ,$$

$$a_i = \langle i|H|i\rangle \,,$$

$$b_i^2 = |H|i\rangle - b_{i-1}|i-1\rangle - a_i|i\rangle|^2 \,.$$

H has to be applied $m-1$ times only instead of m^2 times. In actual computations truncation errors can occur and reorthogonalization can be necessary. As Roomany *et al.*[20] we found no need to do so. Reorthogonalization seems of more importance if the Lanczos method is applied to matrices of order m equaling several thousand, cf. Whitehead *et al.*[13]

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