MAGNON-BOUND STATE RESONANCE IN THE QUASI-1D ANTIFERROMAGNET $RbCoCl_3 \cdot 2H_2O$

E.J. NIJHOF, H. van der VLIST and G.J. GERRITSMA

Twente University of Technology, Department of Applied Physics, P.O. Box 217, 7500 AE Enschede, The Netherlands

Electron spin resonance is studied for 2.6 K < T < 6.4 K at 9 GHz. The absorption peak lies at $H \approx 0$ and has maximum intensity just above T_N. From a realistic spin Hamiltonian we infer that magnon-bound state resonance was observed. The linewidth follows a T^4 law for $T > T_N$.

$RbCoCl_3 \cdot 2H_2O$ (RCC) belongs to a series of canted antiferromagnets that exhibit metamagnetic phase transitions below the 3D ordering temperature ($T_N = 2.97$ K) and a typical quasi-1D behaviour for $T_N < T \lesssim |J/k_B|$ ($J/k_B = -14$ K). The present compound discerns itself, however, in having a different crystal structure (monoclinic), a very low metamagnetic transition field (< 10 mT), and a plane of magnetic ordering (bc'-plane) which does not coincide with a typical crystal plane. In addition, two neighbouring $[Cl_4(H_2O)_2]$-octahedra are canted with respect to each other, having the result that the typical Ising behaviour of Co^{2+} compounds is not observed in RCC.

A detailed crystal-field analysis shows that the fictitious $s = 1/2$ Hamiltonian may be written as the sum of an exchange term $\mathscr{H}^{ex}$, a dipolar term $\mathscr{H}^{dip}$ and a Zeeman-term $\mathscr{H}^{ze}$ [1], where

$$\mathscr{H}^{ex} = -2J\sum_i \left(s_i^z s_{i+1}^z + \eta s_i^y s_{i+1}^y + \epsilon s_i^x s_{i+1}^x \right), \qquad (1)$$

$$\mathscr{H}^{dip} = -\mu_B \mu_0 H^{dip} \sum_i (-1)^i s_i^z, \qquad (2)$$

$$\mathscr{H}^{ze} = -\mu_B\mu_0 \left\{ g^{zz} H^z \sum_i s_i^z + g^{yy} H^y \sum_i s_i^z + g^{xx} H^x \sum_i s_i^x + g^{yz} \left(H^z \sum_i (-1)^i s_i^y + H^y \sum_i (-1)^i s_i^z \right) \right\}. \qquad (3)$$

The z axis is taken along the crystallographic b axis, the y axis points along the projection of the magnetic moment in the ac plane. From this Hamiltonian expressions for high temperature susceptibilities were derived as well as for the magnetic moment. From experimental data crystal field parameters and exchange constant may be obtained leading to the following values for the various terms in the Hamiltonian [1]: $J/k_B = -14.5$ K, $\eta \approx 1$, $\epsilon = 0.6$, $g^{zz} = g^{yy} = 4.6 \pm 0.5$, $g^{xx} = 3.2 \pm 0.4$, $g^{yz} = 1.4 \pm 0.5$, and the dipolar term may include the exchange fields from neighbouring chains. It is seen that the exchange term is far from being Ising-like and that there appear staggered fields in the dipolar and Zeeman terms. If we take $\eta = \epsilon$ and neglect the non-staggered terms of $\mathscr{H}^{ze}$ our antiferromagnetic Hamiltonian can be mapped onto a ferromagnetic Hamiltonian by two successive unitary transformations $U_1 = \Pi' \sigma^z$ and $U_2 = \Pi' \sigma^x$, where the products run over every other spin and the σ's denote the Pauli spin matrices. This results in

$$\mathscr{H} = -2J\sum_i \left\{ s_i^z s_{i+1}^z + \tfrac{1}{2}\epsilon \left(s_i^+ s_{i+1}^+ + s_i^- s_{i+1}^- \right) \right\} + h\sum_i s_i^z, \qquad (4)$$

where h is the total effective (now non-staggered) field. In zero field this Hamiltonian gives rise to n-magnon bound states (n-MBS's). An n-MBS ($n \geq 2$) may be considered as the continuous counterpart of the n-fold spin cluster of an Ising system ($\epsilon = 0$) and evolves towards a state with n decoupled spin waves for a pure Heisenberg chain ($\epsilon = 1$). For small ϵ, the energies form a continuous band around $2|J|$ having a width of $8\epsilon|J|$ at wave number $k = 0$ [2]. For arbitrary ϵ, the dispersion relation is given in terms of elliptic integrals by Johnson et al. [3]. Using Ising basis functions the dispersion relation of eq. (4) was solved by Fogedby and Jensen [4] in terms of the zeros of Bessel functions. The continuous excitation spectrum breaks up into discrete levels that start from the lower band edge with infinite derivative. The $\Delta s^z = 0$ transition frequencies are given by [1]

$$\hbar\omega = 2\epsilon|J| \left[3\pi g^{yz} \mu_B \mu_0 H^{eff} / 2\epsilon|J| \right]^{2/3} \times \left[\left(\kappa + \tfrac{7}{4}\right)^{2/3} - \left(\kappa + \tfrac{3}{4}\right)^{2/3} \right], \qquad (5)$$

and are depicted for $\kappa = 0, 1, 2$ in fig. 1. The effective field H^{eff} is the sum of internal and applied fields along the y axis (c' axis). From susceptibility measurements for $T \gtrsim T_N$ the internal field, due to neighbouring chains in the bc-plane $H^{int} \approx 40$ mT [5]. The non-staggered external-field components of $\mathscr{H}^{ze}$ shift the energy levels an equal amount for all κ [6].

The microwave absorption of RCC was measured using a home-made X-band spectrometer which directly records absorption. In fields up to 3 T all absorption peaks may be combined into one formula

$$y = y^0 \cos^2\theta_{rf} / \left\{ 1 + \left(H\cos\theta_{dc}/\Gamma_0 \right)^2 \right\} \qquad (6)$$

with y^0 = amplitude, Γ_0 = linewidth, θ_{rf} and θ_{dc} are the angles between rf-field, respectively, dc-field and the c'-axis in the ac-plane. Above T_N the maximum of the

0304-8853/86/$03.50

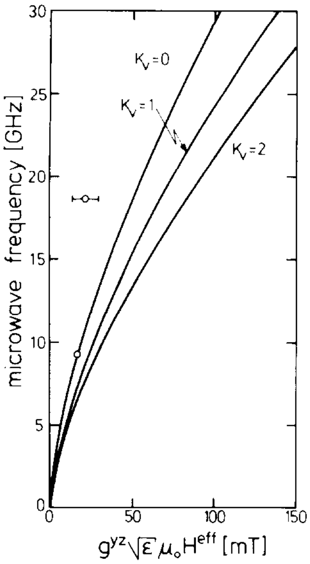

Fig. 1. Lowest three MBS-resonance frequencies versus effective field. Included are two experimental data points for $T = 3.2$ K, $\epsilon = 0.1$ and $H^{eff} = H^{int} = 40$ mT and $H_{dc} = 0$.

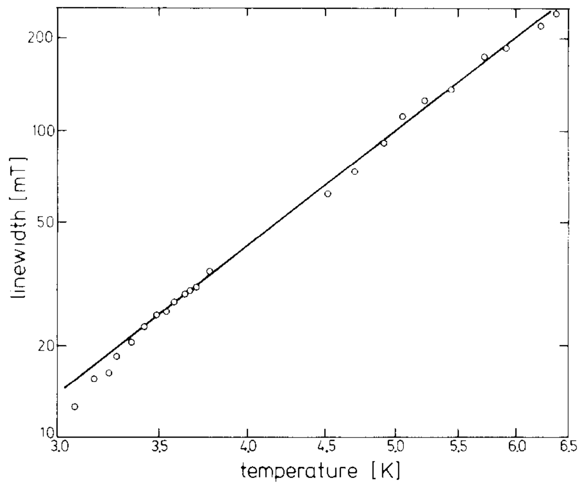

Fig. 3. Linewidth of MBS-resonance versus temperature on double logarithmic scale. ——— $\Gamma \sim T^{3.9}$.

absorption occurs at zero field, whereas somewhat below T_N the peak shifts a little bit (max. 5 mT). The striking angular dependencies of both microwave and external fields can be explained as follows. A component of the microwave field parallel to s_i^z is required for a $\Delta s^z = 0$ transition and this can only be accomplished by the component of the rf-field along the c' direction, assuming that the rf-field rotates in the ac plane. Similarly only the component of the external field parallel to the c' direction shifts the resonance frequencies of the $\Delta s^z = 0$ transitions and hence along the c' axis the observed linewidth is minimal. The calculated curve for $\kappa = 0$ in fig. 1 is drawn through 9 GHz. We also included an experimental point at 18 GHz [7]. For both experimental points the required external field is too small to be distinguished from zero because of the linewidth. The maximum intensity of the absorption is plotted against $1/T$ in fig. 2. In order to observe microwave absorption the MBS's have to be thermally excited and if E is the excitation energy, the intensity should be proportional to $\exp(-E/k_B T)$. The solid line yields $E/k_B = (26 \pm 2)$ K, which is quite close to $2|J|/k_B$. Below T_N, the intensity drops far to rapidly to be explained by thermal excitation of MBS's (dashed line). However, in this case we have a rapid rise of the internal (staggered) field and hence a rapid increase of the resonance frequency thereby taking it out of the experimental region. The linewidth Γ_0 against temperature is depicted in fig. 3. We find for $T > T_N$, $\Gamma_0 \sim T^{3.9}$ (solid line). This may be compared with calculations of antiferromagnetic resonance linewidth ($\epsilon = 1$) based on multimagnon processes [8] yielding T^{α} with $\alpha = 2$ or 4 depending on dimensionality and temperature region, although their results are derived for $T \ll T_N$.

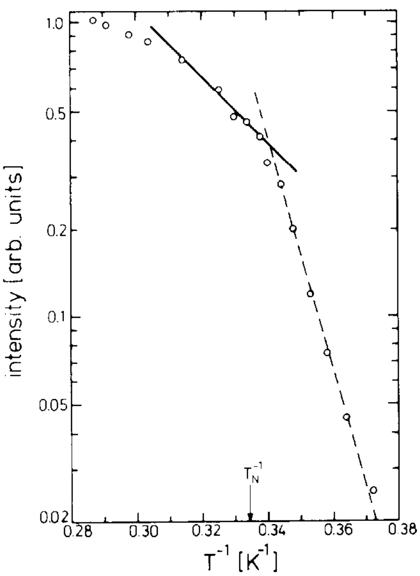

Fig. 2. MBS-resonance intensity versus reciprocal temperature. ——— $E/k_B = 26$ K, - - - $E/k_B = 93$ K.

To the best of knowledge no such theory is available for MBS resonances.

[1] E.J. Nijhof, Thesis, Enschede (1985).
[2] N. Ishimura and H. Shiba, Progr. Theor. Phys. 63 (1980) 743.
[3] J.D. Johnson, S. Krinsky and B.M. McCoy, Phys. Rev. A8 (1973) 2526.
[4] H.C. Fogedby and H.H. Jensen, Phys. Rev. B6 (1976) 3444.
[5] J. Flokstra, G.J. Gerritsma, A.J.W.A. Vermeulen, E. Frikkee and W.J.M. de Jonge, to be published.
[6] K. Adachi, J. Phys. Soc. Japan 50 (1981) 3904.
[7] C.H.W. Swüste, unpublished results.
[8] S.M. Rezende and R.M. White, Phys. Rev. B14 (1976) 2939.