Angular Dependence of the ESR Linewidth in $CuGeO_3$.

Rushana M. Eremina [a,1], M.V.Eremin [b], V.N.Glazkov [c], H.-A. Krug von Nidda [d], A.Loidl [d]

[a] *E.K.Zavoisky Physical Technical Institute, 420029 Kazan, Russia*
[b] *Kazan State University, 420008 Kazan, Russia*
[c] *P.L.Kapitza Institute for Physical Problems, 117334 Moscow, Russia*
[d] *EPV EKM, Institute für Physik, Universität Augsburg, Germany*

Abstract

Analysis of orientation dependencies of ESR linewidth in $CuGeO_3$ taken at different temperatures. Intrinsic anisotropic exchange parameters of copper spins have been determined.

Key words: $CuGeO_3$; ESR; low dimensional structure

1. Introduction

The quasi one dimensional $S = 1/2$ antiferromagnet $CuGeO_3$ is the only inorganic compound demonstrating a spin-Peierls transition at T_{SP}=14.2K [1] up to now. Therefore this compound has attracted a lot of attention during the last decade.

The crystal structure of $CuGeO_3$ is well deteremined [2]. At high temperatures the unit cell contains two formula units (space group $Pbmm$ (D_{2h}^5)). Each copper ion is surrounded by six oxygen ions forming a slightly deformed octahedron [2]. The oxygen octahedra within one unit cell share the apical oxygen. Chains of edge-sharing CuO_6 octahedra aligned along the c-axis determine the quasi one dimensional magnetic properties of the $CuGeO_3$. There are two inequivalent chains which differ by the orientation of the octahedron axis in the (ab)-plane.

The values of the symmetrical Heisenberg intra chain exchange constant is by now well established from the susceptibility measurements and neutron-scattering experiments as J_c=10.4meV(see [2] and ref. therein).

The constants of the possible anisotropic exchange interactions are still not known. Yamada *et al.* [3] have suggested that the antisymmetric anisotropic exchange interaction can describe the temperature dependence of the ESR linewidth. However, this suggestion contradicts to the crystal structure [2]. Later Pilawa [4] argued that the anisotropy of the ESR linewidth in $CuGeO_3$ should be attributed to symmetric anisotropic exchange interactions. However, the nature of the anisotropic exchange interaction in $CuGeO_3$ is still a subject of discussion.

2. Experimental results and discussion.

The ESR measurements were performed with a Bruker ELEXSYS E500CW spectrometer at X-band frequency (9.5 GHz), equipped with a continuous gas-flow cryostat for He (Oxford Instruments).

Above T_{SP} there is one ESR line, below T_{SP} a fine structure of the line appears, which can be attributed to the spin-cluster formation as described in paper [8]. Here we focus our attention to the angular dependence of the ESR linewidth, which is strongly narrowed due to the exchange interaction, at different temperatures above 10K. The ESR-line half-width is determined by [6]

[1] Corresponding author. Present address:E.K.Zavoisky Physical Technical Institute, 420029 Sibirskij trakt 10/7, Kazan, Russia. E-mail: rushana@dionis.kfti.knc.ru

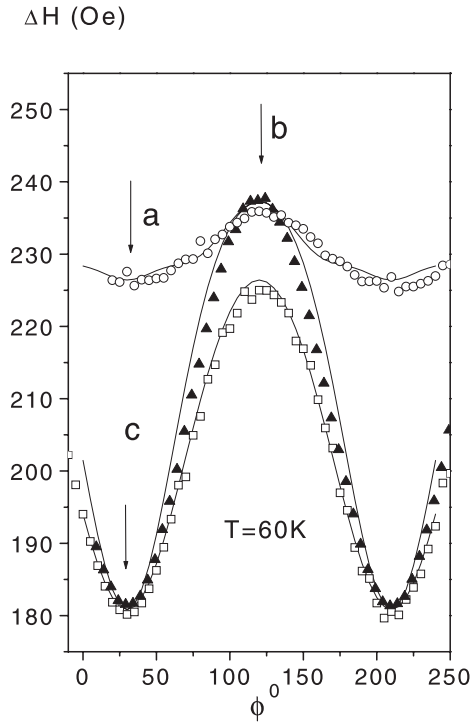

Fig. 1. Angular dependence of the resonance linewidth for the three crystallographic planes (ab), (ac) and (bc) at 60K. The solid lines were obtained by the fit.

$$\Delta H = \frac{M_2}{\omega_{ex}} \tag{1}$$

in high temperature approximation ($J_{ex} \gg T$). Here M_2 is the second moment, caused by anisotropic exchange between the copper spins, ω_{ex} is the so called exchange frequency. Both M_2 and ω_{ex} are temperature dependent due to the effect of antiferromagnetic correlations and saturate at high T. At low temperature ($J_{ex} \ll T$) the relation between ESR linewidth and temperature is predicted to be linear [5] for symmetric anisotropic exchange. In the intermediate temperature regime, from the angular dependencies of the ESR linewidth it is possible to find only the ratio between the anisotropic exchange values. In the case of $CuGeO_3$ the second moment is calculated from the average of the contributions of the two inequivalent chains. In addition, we take into account the contribution to M_2 from the inter-chain interaction.

The symmetric intra-chain anisotropic exchange interaction is written in the form

$$H_1 = \sum [J_{xx} S_i^x S_j^x + J_{yy} S_i^y S_j^y + J_{zz} S_i^z S_j^z] \tag{2}$$

where $J_{xx} + J_{yy} + J_{zz} = 0$. The exchange interaction tensor is diagonal in the coordinate system with the y-axis along the c direction and the z axis perpendicular to the CuO ribbon.

The anisotropic exchange interaction between the chains was included as

$$H_2 = D_{cc} S_c^a S_c^b \tag{3}$$

The second moment M_2 caused by the symmetric anisotropic exchange interaction can be calculated in coordinates with z'- axis along the external magnetic field in high-temperature approximation in analogy to [7].

The angular dependencies of the ESR linewidth where approximated by expressions (2-4) from [7] using (1-3). The g values are $g_{xx} = 2.03$, $g_{yy} = 2.04$, $g_{zz} = 2.325$ in the local coordinate systems of each octahedron. Figure 1 shows the full angular dependence of the resonance linewidth within the three crystallographic planes (ab), (ac) and (bc) at 60K. The relations between the anisotropic exchange parameters are $J_{xx}/J_{zz} = -0.63 \pm 0.02$ and $D_{cc}/J_{zz} = 0.64 \pm 0.02$ in the temperature range 60K-300K. On decreasing temperature near T=25K, the relations between the anisotropic exchange parameters are changed. The contribution of the inter-chain interaction becomes larger $D_{cc}/J_{zz} = 0.87 \pm 0.02$ and the ratio between $J_{xx}/J_{zz} = -0.48 \pm 0.02$ decreases. This is probably connected with the deformation of the crystal structure [2] near 20K.

We note that the relation between the intra-chain anisotropy parameters is almost the same as it was obtained for $LiCuVO_4$. It is remarkable because the chain structures are similar in $LiCuVO_4$ and $CuGeO_3$. Therefore, as in $LiCuVO_4$ [7], we conclude that the unusually large exchange interaction in $CuGeO_3$ originates from the ring geometry of the two oxygen bridge coupling (see [7] and Ref. therein).

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