QUANTUM CRITICAL BEHAVIOUR IN $CuGeO_3$*

S.V. Demishev, A.A. Pronin, N.E. Sluchanko
N.A. Samarin, V.V. Glushkov

General Physics Institute, 38, Vavilov street, 119991 Moscow, Russia

Y. Inagaki, H. Ohta, S. Okubo, Y. Oshima

Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan

and M.M. Markina

Moscow State University, 119899 Moscow, Russia

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From high frequency (up to 450 GHz) ESR and low temperature specific heat measurements we find that insertion of 1% Fe and 2% Co in $CuGeO_3$ matrix smears both spin-Peierls and Néel transitions. For $T < 30$ K we discovered an onset of the power asymptotics of magnetic susceptibility and specific heat characteristic to a quantum critical behaviour.

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1. Introduction

It is known, that disorder in antiferromagnetically interacting spin system can destroy magnetic ordering and give rise to a quantum critical (QC) behaviour [1–3]. For 1D $S = 1/2$ spin chains the ground state becomes gapless and the density of states diverges at $\epsilon = 0 : \rho(\epsilon) \propto |\epsilon|^{-\alpha}$ [1]. As a consequence the temperature dependences of magnetic susceptibility χ and magnetic contribution to specific heat C_m acquire the forms

$$\chi \propto T^{-\alpha}; \qquad c_m \propto T^{1-\alpha}, \tag{1}$$

where $\alpha < 1$. From the theoretical point of view the non-Curie asymptotic behaviour of χ and a power law for C_m reflects the onset of the Griffiths

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phase which thermodynamic properties are controlled by relatively rare spin clusters correlated more strongly than average [2,3].

The possibility of the QC regime in $CuGeO_3$ was considered in [3]. The aim of the present work consists in providing experimental evidence that doping $CuGeO_3$ with magnetic impurities Co and Fe may cause QC behaviour. Details concerning sample preparation and experimental technique can be found elsewhere [4].

2. Experimental results

The key problem in magnetic susceptibility measurements consists in separation of the contribution from disordered Cu^{2+} chains from paramagnetism caused by impurities in $CuGeO_3$ matrix. This difficulty overcomes in ESR experiment, where magnetic impurity in $CuGeO_3$ matrix and Cu^{2+} chains modified by impurity give rise to a different absorption lines [5].

Experimental result for the sample containing 2% of Co is shown in Fig. 1. ESR spectrum is formed by two lines which becomes resolved for

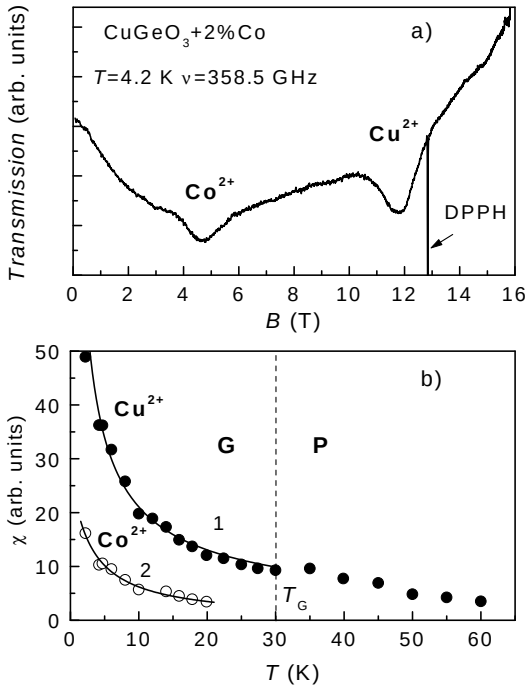

Fig. 1. (a) ESR spectrum for Co-doped sample in geometry $B \parallel a$. Arrow mark reference spectrum for DPPH. (b) χ(T) data for Cu^{2+} chains and Co^{2+} impurity. 1- power law (1) with α=0.69, 2- Curie–Weiss law.

frequency ν>100GHz (Fig. 1(a)). The analysis of the g-factor values together with the frequency and temperature dependences of the linewidth have allowed to associate high-field resonance with $g \approx 2.15$ and low-field resonance with $g \approx 4.7$ with Cu^{2+} chains and Co^{2+} impurity respectively. Data at various temperatures, analogues to presented in Fig. 1(a) was used to calculate $\chi(T)$ for Cu^{2+} and Co^{2+} subsystems (Fig. 1(b)). Susceptibility of the Co-doped spin-Peierls chains for $T < 30$ K follows Eq. (1) with $\alpha = 0.69 \pm 0.04$ (Fig. 1(b) curve 1). At the same time $\chi(T)$ for Co impurity in $CuGeO_3$ matrix is better described by Curie–Weiss law, $\chi \sim 1/(T - \Theta)$, with $\Theta = -2.8$ K (Fig. 1(b) curve 2).

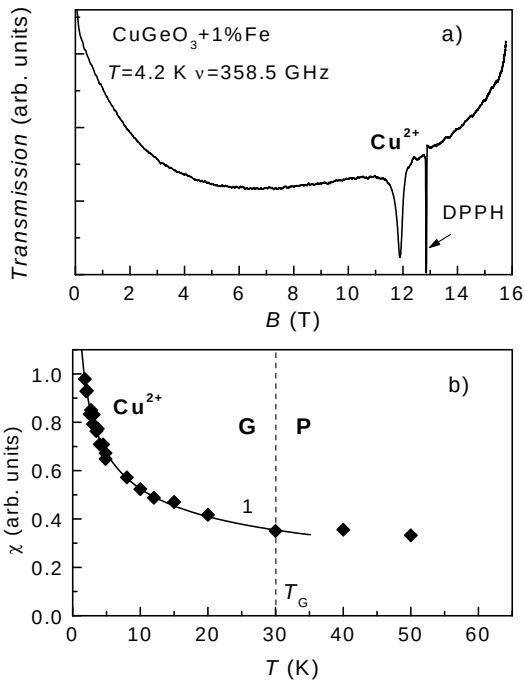

Fig. 2. The same as Fig. 1 for Fe-doped sample. 1- power law (1) with $\alpha = 0.35$.

Results of the similar experiment for the sample containing 1% of Fe are given in Fig. 2. Note that only absorption corresponding to Cu^{2+} chains is observed. As for Co-doped sample the low temperature asymptotic of χ is given by Eq. (1), but the index value is considerably lower: α=0.35±0.03 (Fig. 2(b) curve 1). In addition for the Fe-doped sample we find [4] that magnetic contribution c_m obeys power law with the index α=0.37±0.03 which coincide within experimental error with the value obtained from magnetic susceptibility.

3. Discussion

Data in Fig. 1(b) and Fig. 2(b) suggests that in the samples studied spin-Peierls transition is already damped. Specific heat data for $CuGeO_3$:Fe also show no spin-Peierls transition [4]. It is also possible to exclude Neel ordering expected for doped $CuGeO_3$ [1] as long as $\chi(T)$ have no kinks and no magnetoabsorption modes corresponding to antiferromagnetic resonance have been observed. At the same time the presence of the disorder in magnetic subsystem follows from the strong broadening of the ESR line with respect to the pure crystal (Figs 1,2 and [4]). In this situation an observation of the power asymptotics for Cu^{2+} chains agrees with the QC behaviour. In this model the index α in Eq. (1) is not universal and depends on the distribution function of the spin clusters [2,3] that may explain difference between Fe and Co-doped crystals. Considering the formation of the Griffiths phase at low temperatures it is possible to estimate the value of characteristic temperature $T_G = 30$ K at which transition from paramagnetic to (P) to a Griffiths (G) phase is expected (Figs 1,2). However a further experiments at very low temperatures required to check proposed explanation base on QC regime.

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