Quantum critical point in CuGeO$_3$ doped with magnetic impurities

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Abstract

Using high frequency (up to 450 GHz) ESR and low temperature specific heat measurements we find that insertion of 1% Fe and 2% Co damps spin-Peierls and Neel transitions and for $T < 30$K may give rise to onset of a quantum critical behaviour characteristic for a Griffiths phase.

Key words: CuGeO$_3$; quantum critical phenomena; ESR

Most of the available data for doped inorganic spin-Peierls compound CuGeO$_3$ correspond to the limit of weak disorder when density of states have a pseudo-gap [1,2]. In the opposite case of a strong disorder the situation is expected to change dramatically: the ground state of CuGeO$_3$ should be a Griffiths phase (GP) which thermodynamic properties are controlled by relatively rare spin clusters correlated more strongly than average [3,4]. In this case density of states becomes gapless and diverges at $\epsilon = 0$: $\rho(\epsilon) \propto |\epsilon|^{-\alpha}$ [1]. As a consequence the temperature dependences of magnetic susceptibility $\chi$ and magnetic contribution to specific heat $c_m$ for Cu$^{2+}$ spin-Peierls chains acquire the forms [3,4]

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

where $\alpha < 1$. As long as $T = 0$ becomes singular or critical point, the aforementioned situation may be described in terms of quantum critical (QC) behaviour [3].

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The experimental information about feasibility of the QC point in CuGeO$_3$ is very limited [5]. The aim of the present work consists in providing possible evidence of onset of the QC point in CuGeO$_3$ doped with magnetic impurities Co and Fe.

Single crystals of CuGeO$_3$ doped with 1% of Fe (S=1) and 2% of Co (S=3/2) impurities have been grown using self-flux method. The quality of crystals was controlled by X-ray and Raman scattering data [6]. Magnetic properties of the samples in the temperature range 1.7-300K have been studied by high (up to 450 GHz) frequency ESR technique [2] for $B\parallel a$ geometry. Specific heat for the temperature interval 6-20K was studied with the help of low temperature small sample relaxation calorimeter.

Doping of CuGeO$_3$ with magnetic impurities causes (i) rapid damping of the spin-Peierls transition and (ii) onset of a strong paramagnetic background [7]. Therefore the deriving of the effects of doping on Cu$^{2+}$ S=1/2 chains in convenient magnetisation measurements requires *ad hoc* assumptions about background behaviour [7]. This difficulty overcomes in ESR experiment, where magnetic impurity in CuGeO$_3$ matrix and Cu$^{2+}$ chains modified by impurity gives rise to a different absorption lines [8].

Typical magnetoabsorption spectra for CuGeO$_3$:Co and CuGeO$_3$:Fe at helium temperatures are shown in Fig.1. For the Co-doped sample spectrum is formed by two broad lines which becomes resolved for frequencies $\nu>100$ GHz. The spectrum of Fe-doped sample is characterised by a single resonance. The reso-

![Fig. 1. Magnetoabsorption spectra for the samples studied.](image)
nant fields $B_{res}$ for all modes presented in Fig.1 are found to vary linearly with frequency $B_{res} \sim \nu$. This result suggests that the observed magnetoabsorption features correspond to different ESR modes rather than coexistence of the ESR and antiferromagnetic resonance [1].

The analysis of the $g$-factor values together with the frequency and temperature dependences of the linewidth [2] have allowed to attribute the high field resonances with $g \approx 2.15$ to ESR on disordered Cu$^{2+}$ chains and the low field resonance with $g \approx 4.7$ for CuGeO$_3$:Co to ESR on Co$^{2+}$ impurity in CuGeO$_3$ matrix (Fig.1). A strong $g$-factor renormalisation for the Co-doped sample may reflect formation of the impurity spin clusters [8].

As long as the integrated intensity of the ESR line in the region of linear response is proportional to magnetic susceptibility it was possible to subtract contributions of Cu$^{2+}$ chains and Co$^{2+}$ impurities unambiguously and find $\chi(T)$ dependence for both contributions from the spectra taken at different temperatures (Fig.2). The correctness of the $\chi(T)$ evaluation from ESR spectra has been checked by measurements of the static susceptibility for Co-doped sample by vibrating sample magnetometer. We found that sum of $\chi(T)$ for Cu$^{2+}$ and Co$^{2+}$ resonances represents the integral static susceptibility of the sample within the error of less than 5%.

Quantitative analysis of $\chi(T)$ data for doped Cu$^{2+}$ chains indicates that Eq.(1) holds for $T < 30K$ (curves 1 and 2 in Fig.2) and the power law provides reasonable description of experimental data. The best fit was obtained using index values $\alpha = 0.36 \pm 0.03$ and $\alpha = 0.69 \pm 0.04$ for Fe-doped and Co-doped crystals respectively. In contrast to the magnetic properties of Cu$^{2+}$ chains, the susceptibility for Co$^{2+}$ impurity is better fitted by Curie-Weiss law, $\chi \sim 1/(T-\Theta)$, with characteristic temperature $\Theta = -2.8K$ (Fig.2, curve 3) corresponding to antiferromagnetically interacting Co$^{2+}$ impurities. For doped Cu$^{2+}$ chains it is visible that for magnetic impurities studied spin-Peierls and Neel transitions are already damped.

The observation of the power asymptotics of $\chi(T)$ in the absence of any type of long range magnetic order for the wide range $1.7 < T < 30K$ (where temperature varies more than 15 times) agrees with the theoretical predictions [3,4] for the QC point. An additional argument in favour of proposed interpretation follow from specific heat data for CuGeO$_3$:Fe. It was found [2] that magnetic contribution $c_m$ follows power law with the index $\alpha = 0.37 \pm 0.03$ which coincide within experimental error with the value obtained from magnetic susceptibility.

In conclusion, we provide possible experimental evidence that doping of CuGeO$_3$ with magnetic impurities Co and Fe induces a strong disorder limit and low temperature $T < 30K$ ground state may become a GP. A magnetic structure
Fig. 2. Temperature dependence of magnetic susceptibility for doped Cu$^{2+}$ chains and Co$^{2+}$ impurity. $\chi(T)$ data suggest a transition from paramagnetic (P) to Griffiths (G) phase at $T=30$K (see text for details).

of the clusters in GP, including the possibility of the predicted in [9] random dimer phase, is a subject of future investigations.

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References

[1] M.Mostovoy et al., Phys. Rev. B 58 (1998) 8190.
[2] S.V.Demishev et al., cond-mat/0110177.
[3] A.Rosch, in: Abstracts of LT22, Helsinki (1999) 389.
[4] D.Fisher, Phys. Rev. B 50 (1994) 3799.
[5] K.Manabe et al., Phys. Rev. B 58 (1998) R575.
[6] S.V.Demishev et al., JETP Letters 73 (2001) 31.
[7] P.E.Anderson et al., Phys. Rev. B 56 (1997) 11014.
[8] V.N.Glazkov et al., J.Phys.: Cond. Mat. 10 (1998) 7879.
[9] H. Fabrizio, R. Mellin, Phys. Rev. Lett. 78 (1997) 3382.