

Quantum critical point in CuGeO₃ 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 $T < 30\text{K}$ gives rise to onset of a quantum critical behaviour characteristic for a random dimer Griffiths phase.

Key words: CuGeO₃; quantum critical phenomema; ESR

Most of the available data for doped inorganic spin-Peierls compound CuGeO₃ 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 CuGeO₃ should be a Griffiths phase (GP) which thermodynamic properties are controlled by relatively rare spin clusters correlated more strongly than average [3,4]. In the absence of both long range spin-Peierls and antiferromagnetic orders 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 χ and magnetic contribution to specific heat c_m for Cu²⁺ spin-Peierls chains acquire the forms [3,4]

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

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

The experimental information about feasibility of the QC point in CuGeO₃ is very limited [5]. The aim of the present work consists in providing possible evi-

dence of onset of the QC point in CuGeO₃ doped with magnetic impurities Co and Fe.

Single crystals of CuGeO₃ 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; the actual contents of dopant in the sample was controlled by chemical analysis [6]. Magnetic properties of the samples in the temperature range 1.8-300K have been studied by high (up to 450 GHz) frequency ESR technique [2], ESR magnetoabsorption spectra have

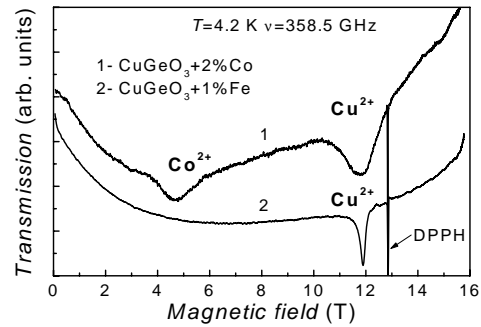


Fig. 1. Magnetoabsorption spectra for the samples studied.

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been measured for $\mathbf{B} \parallel \mathbf{a}$ geometry. Together with the sample spectrum a reference spectrum for DPPH was recorded. 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 $\text{CuGeO}_3\text{:Co}$ and $\text{CuGeO}_3\text{:Fe}$ 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 resonant 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 [1,9].

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 $\text{CuGeO}_3\text{:Co}$ 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 contribution of Cu^{2+} chains unambiguously and find $\chi(T)$ dependence from the spectra taken at different temperatures (Fig.2). We also calculated $\chi(T)$ for Co^{2+} impurity (Fig.2). It is visible that for magnetic impurities studied spin-Peierls transition is already damped and Neel ordering is missing up to 1.8K.

Quantitative analysis of $\chi(T)$ data for doped Cu^{2+} chains indicates that Eq.(1) holds for $T < 30\text{K}$ (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 $\Theta = -2.8\text{K}$ (Fig.2, curve 3) corresponding to antiferromagnetically interacting Co^{2+} impurities.

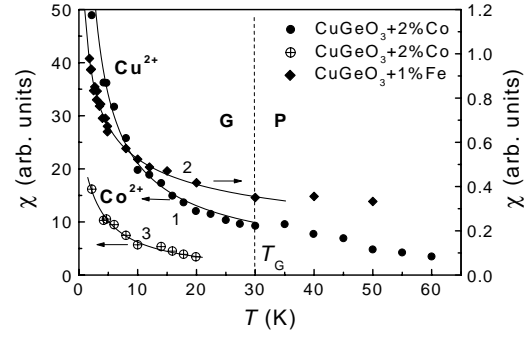


Fig. 2. Temperature dependence of magnetic susceptibility for doped Cu^{2+} chains and Co^{2+} impurity. $\chi(T)$ data suggest a transition from paramagnetic (P) to Griffiths (G) phase at $T = 30\text{K}$ (see text for details).

The observation of the power asymptotics of $\chi(T)$ in the absence of any type of long range magnetic order for the wide range $1.7 < T < 30\text{K}$ (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 $\text{CuGeO}_3\text{:Fe}$. 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 $T < 30\text{K}$ ground state becomes gapless random dimer GP [4]. Further studies of the magnetic ordering of these solids at very low temperatures may be prospective.

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