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Angaben zur Veröffentlichung / Publication details:

Glazkov, V. N., A. I. Smirnov, Hans-Albrecht Krug von Nidda, Alois Loidl, T. Masuda, and K. Uchinokura. 2004. "ESR study of the impurity-induced antiferromagnetic ordering in the diluted spin-Peierls magnet CuGeO₃." *Journal of Magnetism and Magnetic Materials* 272-276 (Supplement): E695–96. <https://doi.org/10.1016/j.jmmm.2003.12.350>.

ESR study of the impurity-induced antiferromagnetic ordering in the diluted spin-Peierls magnet CuGeO_3

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The spin-gap systems like spin-Peierls or Haldane magnets, containing one-dimensional spin chains with antiferromagnetic (AFM) exchange, have a singlet spin-disordered ground state and the vanishing magnetic susceptibility at $T \rightarrow 0$. Impurities locally destroy the nonmagnetic state and result in the appearing of areas of AFM-correlated spins (a kind of spin clusters), located near impurities. At low temperatures, the locally ordered areas should overlap and form the ordered phase due to the interchain exchange.

ESR is a convenient tool to probe AFM order formation, since AFM and paramagnetic resonance signals can be easily resolved. In our previous ESR-study of Mg-diluted spin-Peierls magnet CuGeO_3 we have found a microscopic phase separation [1]. It was revealed at low doping level as the coexistence of the AFM and paramagnetic resonance signals below the transition temperature T_N . Here we study the influence of the magnetic field and of the impurity concentration

on the paramagnetic fraction below T_N and the structure of the phase above T_N .

The paramagnetic fraction was measured as relative intensity of the paramagnetic spectral component and appeared to be dependent on the magnetic field and concentration (Fig. 1). The paramagnetic phase fraction increases with increasing magnetic field, and decreases with increasing impurity concentration.

At temperatures $T \gtrsim 2T_N$ the ESR line has a single-Lorentzian form. At lower temperatures $T_N < T \lesssim 2T_N$, the ESR line is a sum of the two Lorentzian components with the same resonance field, but different linewidths (Fig. 2). First of these components broadens as temperature decreases, and, at the transition point, it transforms into the AFM resonance line. The second component demonstrates no changes at T_N and remains in the same resonance field providing the paramagnetic component of the ESR line below T_N . Thus, two kinds of ESR signals are present also above T_N .

These findings are explained considering clusters of the staggered magnetization formed near the impurity [1]. The magnitude of the staggered magnetization

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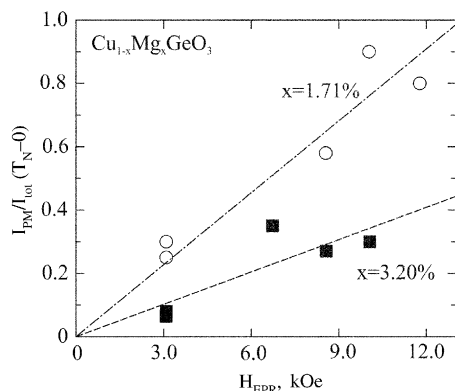


Fig. 1. Paramagnetic component intensity, approximated to the Neel point, as a function of magnetic field.

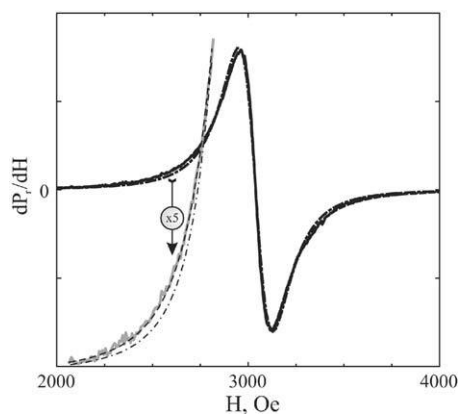


Fig. 2. ESR line in the $\text{Cu}_{1-x}\text{Mg}_x\text{GeO}_3$ at $T > T_N$, $x = 1.71\%$, $f = 9.5$ GHz, $\mathbf{H} \parallel b$, $T = 2.6$ K, $T_N = 2.25$ K. Dashed line — two Lorentzian fit, dash-dotted line — one Lorentzian fit. Grey solid line — five-fold amplified experimental curve.

decays with the distance from the impurity ion and, at some distance, AFM correlations are destroyed by the thermal fluctuations. The size of the coherent order area increases with decreasing temperature.

At $T \gg T_N$ areas of local order do not overlap. They provide the paramagnetic resonance signal due to their net magnetic moment. ESR line consists of only one

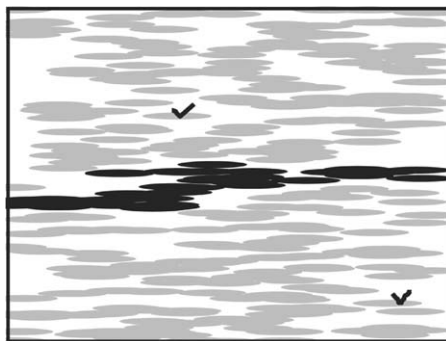


Fig. 3. 2D modeling of the impurity induced AFM ordering. Grey — areas of the coherent AFM order, white — dimerized matrix, black — macroscopic area with coherent AFM order parameter. Ticks mark isolated clusters.

component. As temperature decreases areas of local order merge, and conglomerates with coherent AFM order are formed (Fig. 3). Due to the random distribution of the impurities, volumes of these conglomerates are different. Critical fluctuations give rise to the increase of the ESR linewidth of the large conglomerates. At the Neel point these conglomerates percolate through the macroscopic distance, and corresponding ESR signal transforms into the AFM resonance signal. At the same time, certain isolated clusters remain, giving rise to the narrow paramagnetic spectral component both above and below T_N . Thus, observation of the two components of the paramagnetic line above T_N confirms the existence of local AFM order areas of different sizes.

The coherent magnetic order should be destroyed by the magnetic field, since each cluster has a net magnetic moment μ_B . This leads to the increase of the paramagnetic fraction with increasing magnetic field in accordance with the experimental observations.

References

- [1] V. Glazkov, et al., Phys. Rev. B 65 (2002) 144427.