Microscopic Magnetic Phase Separation at the Impurity Stimulated Antiferromagnetic Ordering of two Spin-Gap Magnets

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Abstract

The ESR spectra of spin-Peierls crystal $CuGeO_3$ and Haldane magnet $PbNi_2V_2O_8$ doped with Mg revealed the coexistence of paramagnetic and antiferromagnetic spin resonance modes below the points of the impurity induced ordering. The ordering is known to be due to the restoring of the AFM staggered magnetization near impurities. The surprising paramagnetic component observed at low doping indicates a nonuniform structure of the ordered phase with a portion of clusters of staggered magnetization which are isolated from the merged clusters by slightly disturbed spin-gap matrix. Isolated clusters provide paramagnetic signals due to their net spin.

Key words: low-dimensional magnetism, spin-gap, impurity induced magnetic ordering

1. Introduction

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. 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. The amplitude of the local AFM order decays exponentially at moving away from an impurity, thus at the wings of the clusters the correlations are destroyed by thermal fluctuations. At low temperatures the locally ordered areas should overlap and form the long-range ordered phase due to the interchain exchange. This impurity-induced AFM-ordering was observed experimentally in the spin-Peierls compound $CuGeO_3$ (see, e.g. Ref. [1]) and in the Haldane magnet $PbNi_2V_2O_8$ [2]. In the present paper we review a wide-range (9-120 GHz) magnetic resonance study of the impurity induced ordering in both systems. Collecting the spectra of the electron spin resonance at cooling the samples through the Néel temperature we follow the transition to the AFM-state and, due to the spectral separation of the paramagnetic and antiferromagnetic resonance (AFMR) signals we can search the transformation from the paramagnetic to the spatially nonunform AFM phase.

2. Experimental results and discussion

We studied the single crystals of $Cu_{1-x}Mg_xGeO_3$ which are well ascribed to the $x-T$ phase diagram reported in Ref. [1]. The typical AFM crystals usually demonstrate a shift of the magnetic resonance line from the paramagnetic resonance position to the AFMR point below the Néel temperature. We found, that at $x \geq 0.04$ the spectrum of the magnetic resonance of

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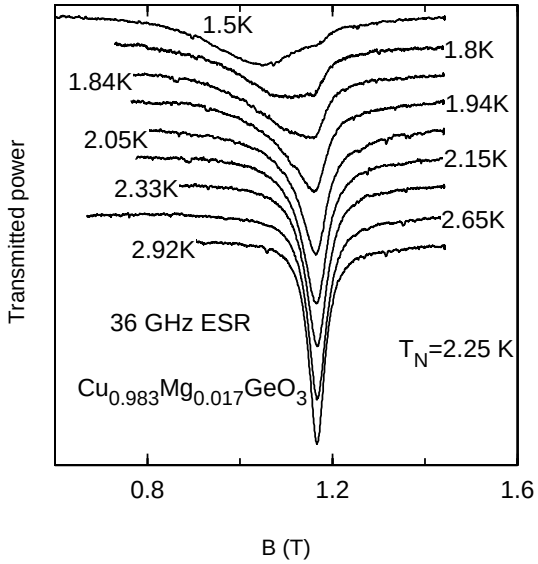

Fig. 1. The temperature evolution of the ESR-absorption in the doped spin-Peierls crystal at cooling through the Néel temperature.

the doped spin-Peierls compound $CuGeO_3$ behaves like that of a typical AFM. However, at low concentrations (x =0.017, 0.032), there is a coexistence of an AFMR signal and of a paramagnetic resonance signal, as one can see on Fig. 1.

The ceramic samples of doped Haldane system $Pb(Ni_{1-y}Mg_y)_2V_2O_8$ have the maximum of the Néel temperature of about 3.5 K at y =0.04 (see Ref. [2]). Above this temperature, for the whole concentration range $0.01 \leq y \leq 0.06$, we observed the paramagnetic resonance signal provided by the impurities, with g-factor near 2.1 and with frequency tending to zero in zero field. Below the Néel points there is a strong distortion of the ESR line corresponding to the absorption band in a polycrystalline antiferromagnetic sample. There is a difference in the temperature-evolution of magnetic resonance signals of the samples with $y \leq 0.02$ and of the samples with $y \geq$0.04. A spectral density at the paramagnetic resonance field survives below the Néel point for $y \leq$0.02, however there is no absorption at the paramagnetic resonance field for $y \geq$0.04. These observations also reveal the coexistence of paramagnetic and antiferromagnetic resonance signals at low concentrations of impurities analogous to the experiments with doped $CuGeO_3$ described above. The small width of the phase transitions to the Néel state (not larger than 0.1 K in both systems) excludes the presence of the paramagnetic phase due to the trivial macroscopic inhomogeneity of the Néel temperature along the sample. The AFM fraction extrapolated to the Néel point was estimated for $Cu_{1-x}Mg_xGeO_3$, it is concentration-dependent and is about 0.3 for x =0.017.

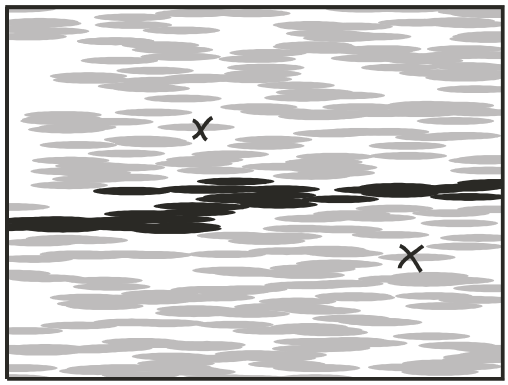

Fig. 2. Geometric model of the microscopic phase separation at the antiferromagnetic phase transition stimulated by the impurities in a spin-gap crystals. The impurities are distributed as random and are centers of areas of the local antiferromagnetic areas marked with grey color. The horizontal size of the figure is 600 interspin distances along spin chains.

The surprising paramagnetic component of the magnetic resonance spectrum, found below the Néel point, indicates a microscopically nonuniform phase with areas providing AFMR signals, areas providing paramagnetic response, and, probably, with "silent" spin-gap areas. We interpret this phase separation as a natural coexistence of clusters of staggered magnetization, touching each other and forming AFM areas, and of single clusters which are isolated from AFM areas by the residual of the spin-gap matrix. Isolated clusters provide PM signals due to their net spin. Isolating areas are "silent" (giving a negligible magnetic response). The described structure is confirmed by a simplified two-dimensional modeling considering the destruction of AFM correlations at the periphery of a cluster at finite temperature and is shown in Fig. 2. Areas of a coherent AFM order are gray, two of isolated clusters are marked by crosses, black area is the largest continuous AFM region percolating through the sample . White space is the spin-gap matrix.

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