

## NOVEL MAGNETISM IN SPIN GAP SYSTEM

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### INTRODUCTION

Most of magnetic properties of conventional magnets can be understood in terms of the classical vector spin model. However, recent development of new quantum magnets and high-field experiments at low temperatures revealed that some systems exhibit many-body effects characteristic of quantum particle system. Such behavior can be observed in spin dimer system, where pairs of ions with spin  $S=1/2$  are coupled antiferromagnetically to form non magnetic singlet state at low temperatures.

### FIELD-INDUCED MAGNETIC ORDERING IN $\text{TiCuCl}_3$

Quantum phase transitions in coupled antiferromagnetic spin dimer systems are of current interest. These systems often have the gapped singlet ground state. In a magnetic field higher than the gap field  $\tilde{A}/g\mu_B$ , created triplet magnons ( $S_z=+1$ ) can hop to neighboring dimers and interact with one another due to the transverse and longitudinal components of the interdimer interaction, respectively. Consequently, the system can be mapped onto an interacting boson model [1]. When the repulsive interaction due to antiferromagnetic interdimer interactions is dominant, magnons can form the superstructure accompanied by the magnetization plateau as observed in  $\text{SrCu}_2(\text{BO}_3)_2$  [2]. On the other hand, when the hopping term is dominant, magnons can undergo Bose-Einstein Condensation (BEC) with the transverse magnetic ordering [3].

$\text{TiCuCl}_3$  is a three-dimensionally coupled spin dimer system with a gapped singlet ground state. The gap  $\tilde{A}$  ( $=0.66$  meV) is significantly smaller than the intradimer interaction  $J$  ( $=5.68$  meV) due to strong interdimer interactions. The small spin gap enables us to observe the magnetic ordering using superconducting magnet. The field-induced magnetic ordering and the excitations in ordered state have been extensively studied by various techniques [4,5,6,7]. The results obtained were in accordance with the magnon BEC model [3,8].

### RANDOMNESS EFFECT ON THE FIELD-INDUCED MAGNETIC ORDERING IN $\text{Ti}_{1-x}\text{K}_x\text{CuCl}_3$

The strong intradimer interaction  $J$  corresponds to the local potential of magnons. Since the values of  $J$  in  $\text{TiCuCl}_3$  and isostructural  $\text{KCuCl}_3$  ( $J=4.34$  meV) are different, we can expect that the partial  $\text{K}^+$  ion substitution for  $\text{Ti}^+$  ions produces the random local potential and the magnitude of randomness varies with the potassium concentration  $x$ . The effect of randomness on the field-induced magnetic ordering was investigated through magnetization and specific heat measurements in  $\text{Ti}_{1-x}\text{K}_x\text{CuCl}_3$ . Well-defined phase transitions were observed. For finite  $x$ , the singlet ground state for  $H < H_c$  turns into the magnetic state with the finite susceptibility due to randomness. However, the magnetic ordering was not detected down to 0.45 K for  $H < H_c$ . This behavior indicates that magnons are localized, and is consistent with the nature of the Bose glass phase [9], in which bosons are localized due to randomness, but there is no gap, so that the compressibility is finite. With increasing  $x$ , the ordering temperature is fairly suppressed. The phase boundary for temperature ( $T$ ) versus magnetic field ( $H$ ) diagram cannot be described by the power law with a single exponent and is described rather well by a linear function of  $T$  at low temperatures. This behavior is in contrast with the power law  $T(H) \propto (H - H_c)^\phi$  with  $\phi \approx 2.0$  [10] in  $\text{TiCuCl}_3$  and interpreted as the crossover to the power law with  $\phi < 1$ . These results suggest that the field-induced magnetic ordering in  $\text{Ti}_{1-x}\text{K}_x\text{CuCl}_3$  belongs to the universality class of the Bose glass-superfluid transition as discussed by Fisher et al. [9].

### PRESSURE-INDUCED MAGNETIC ORDERING IN $\text{TiCuCl}_3$

The spin gap is reduced under the hydrostatic pressure and closes completely at  $P_c \approx 0.07$  GPa [11]. The magnetization at  $P = P_c$  is proportional not to  $H$ , but to  $H^3$  in the low field region. The magnetic phase diagram in the pressure-induced ordered phase was determined

for  $P \leq 0.8$  GPa. The spin flop transition was observed at  $H \approx 0.7$  T for  $H$  parallel to the  $[2,0,1]$  direction. Therefore the easy-axis is close to the  $[2,0,1]$  direction. The ordering temperature at  $P \approx 0.8$  GPa is  $T_N = 11$  K. The Bragg reflections indicative of the magnetic ordering were observed at  $Q=(h, 0, l)$  with integer  $h$  and odd  $l$ , which are equivalent to those for the lowest magnetic excitation at zero pressure and those for the magnetic Bragg peaks indicative of the field-induced magnetic ordering for  $H/b$  [12]. Polarized neutron scattering experiments revealed the successive magnetic ordering at  $P \approx 1.5$  GPa.

### IMPURITY-INDUCED AND FIELD-INDUCED MAGNETIC ORDERINGS IN $\text{TiCu}_{1-x}\text{Mg}_x\text{Cl}_3$

When nonmagnetic ions are substituted for magnetic ions in a spin gap system, the singlet ground state is disturbed so that staggered moments are induced around the impurities. If the induced moments interact through effective exchange interactions, which are mediated by intermediate singlet spins, three-dimensional long range order can arise. Such impurity-induced antiferromagnetic ordering was also observed in  $\text{TiCu}_{1-x}\text{Mg}_x\text{Cl}_3$  [13]. The magnetic structure and the magnetic excitations in the impurity-induced phase were investigated by neutron scattering experiments. It was found that the impurity-induced antiferromagnetic ordering coexists with the spin gap and that the magnitude of gap increases steeply below  $T_N$  upon decreasing temperature [14]. The magnetic ordering under external field was investigated by specific heat measurements. Preliminary data show that the impurity-induced ordered phase and the field-induced ordered phase are continuously connected without any transition separating them. This implies that both phases are identical.

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