Localization of Spin Triplets in Quantum Dimer System $Tl_{1-x}K_xCuCl_3$

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The effect of exchange randomness on the ground state and the field-induced magnetic ordering was investigated through specific heat and magnetization measurements in $Tl_{1-x}K_xCuCl_3$ with $x \leq 0.22$. The isostructural parent compounds $TlCuCl_3$ and $KCuCl_3$ are coupled spin dimer systems with a gapped singlet ground state and their field-induced antiferromagnetic ordering is described by the Bose condensation of spin triplets. Due to exchange randomness, the low-field singlet ground state turns into the magnetic state with finite susceptibility. However, the gap for the triplet excitation still remains. Well-defined field-induced phase transitions were observed in $Tl_{1-x}K_xCuCl_3$. The relation between transition field $H_N(T)$ and temperature is represented by the power law $H_N(T) - H_c \propto T^{\phi}$. Systematic decrease of the critical exponent ϕ with x is observed. These properties are discussed in connection with Bose gas in the presence of random potential.

§1. Introduction

$TlCuCl_3$ and $KCuCl_3$ have the same monoclinic crystal structure composed of chemical dimer Cu_2Cl_6.[1),2)] Two Cu^{2+} ions in the chemical dimer are strongly coupled by the antiferromagnetic exchange interaction J to form an $S = 1/2$ spin dimer; $J/k_B = 65.9$ K and 50.4 K for $TlCuCl_3$ and $KCuCl_3$,[3)-5)] respectively. Their magnetic ground states are spin singlets with excitation gaps Δ/k_B of 7.5 K and 31 K,[6)-8)] respectively. The gaps originate from the strong intradimer interaction J. The neighboring spin dimers couple via three-dimensional interdimer exchange interactions. The spin triplets called magnons (or triplons) can hop to neighboring dimers due to the transverse component of the interdimer interaction. Consequently, the magnon excitations have dispersion and the lowest excitation corresponding to the gap becomes smaller than J.[3)-5),9)] The magnons interact with one another due to the longitudinal components of the interdimer exchange interaction. Hence, the system can be represented as a system of interacting bosons.[10)]

In a finite magnetic field, $|1, 1\rangle$ magnons, whose energy decreases linearly with magnetic field, play an important role. When the hopping of magnon is dominant, as in $TlCuCl_3$ and $KCuCl_3$, $|1, 1\rangle$ magnons can undergo Bose-Einstein condensation (BEC) in a magnetic field higher than the critical field H_c corresponding to the gap. This leads to field-induced transverse magnetic ordering.[11)] Field-induced magnetic ordering in $TlCuCl_3$ has been extensively studied by various techniques.[2),7),12)-14)] The results obtained were in accordance with the magnon BEC model.[11),15),16)]

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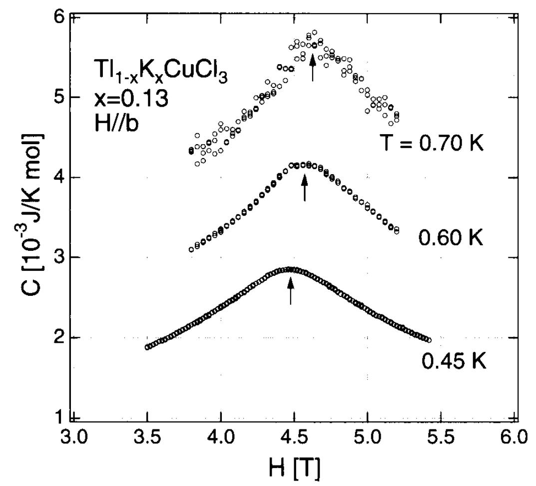

Fig. 1. Field dependence of the specific heat C in $Tl_{1-x}K_xCuCl_3$ with $x = 0.13$ at various temperatures for $H \parallel b$. Arrows denote the transition field $H_N(T)$.

The dominant intradimer interaction J corresponds to the local potential of magnons. Since the values of J for $TlCuCl_3$ and $KCuCl_3$ are different, the partial K^+ ion substitution for Tl^+ ions should produce random local potential. Fisher et al.[17] theoretically discussed the behavior of lattice bosons in random potential, and argued that a new Bose glass phase exists at $T = 0$ in addition to superfluid and Mott insulating phases, which correspond to the field-induced ordered phase and the gapped singlet state, respectively, in the present spin system. In the Bose glass phase, bosons are localized due to randomness, but there is no gap and the compressibility is finite. Fisher et al. showed that the superfluid transition occurs only from the Bose glass phase. They also predicted that the critical behavior near $T = 0$ is different from that for the system without disorder. Thus, we can expect the emergence of the Bose glass phase and the new critical behavior in $Tl_{1-x}K_xCuCl_3$. With this reasoning, we performed magnetization and specific heat measurements on $Tl_{1-x}K_xCuCl_3$ in magnetic fields.

§2. Experimental details

Single crystals of $Tl_{1-x}K_xCuCl_3$ with $x \leq 0.22$ were grown from a melt by the Bridgman method. The details of preparation were reported in reference.[18] Potassium concentration x was determined by emission spectrochemical analysis. Magnetization was measured down to 1.8 K in magnetic fields up to 7 T, using a SQUID magnetometer (Quantum Design MPMS XL). The specific heat was measured down to 0.45 K, using a Physical Property Measurement System (Quantum Design PPMS) by the relaxation method.

§3. Results and discussion

We measured the temperature dependence of the specific heat in $Tl_{1-x}K_xCuCl_3$ at various magnetic fields. At zero field, no anomaly indicative of magnetic ordering is observed down to 0.45 K. For $H > 5$ T, a cusplike anomaly due to magnetic ordering was observed.[19] The anomaly in the present mixed systems is as sharp as that in $TlCuCl_3$.[12] The phase transition is well-defined for $T > 2$ K. The specific heat anomaly below 2 K is so small that it is hard to distinguish the ordering temperature. Then, we measured the field dependence of the specific heat for $T \leq 2$ K. Some examples of the measurements for $x = 0.13$ are shown in Fig. 1. The specific heat displays a clear cusplike anomaly, to which we assign the transition field $H_N(T)$. No

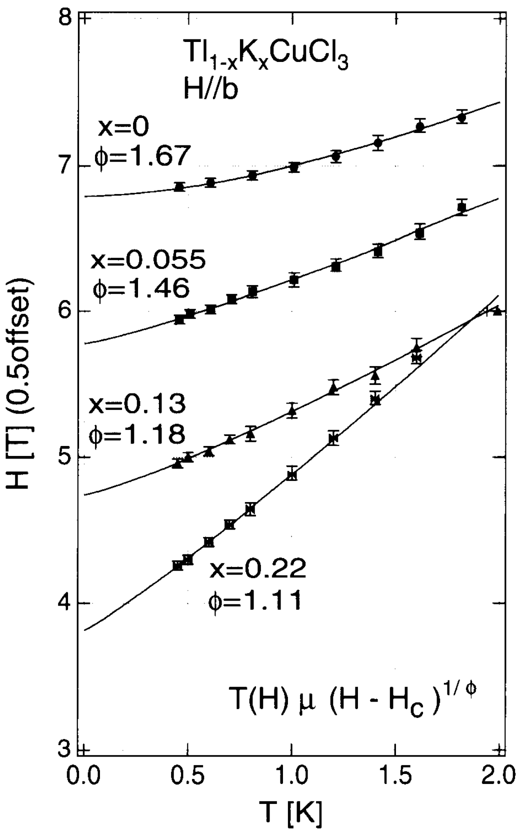

Fig. 2. Low-temperature magnetic diagram in $Tl_{1-x}K_xCuCl_3$ for $H \parallel b$. The values of the external field H are shifted upward consecutively by 0.5 T with decreasing x. Solid lines denote the fits by the power law with exponents ϕ shown in Figure.

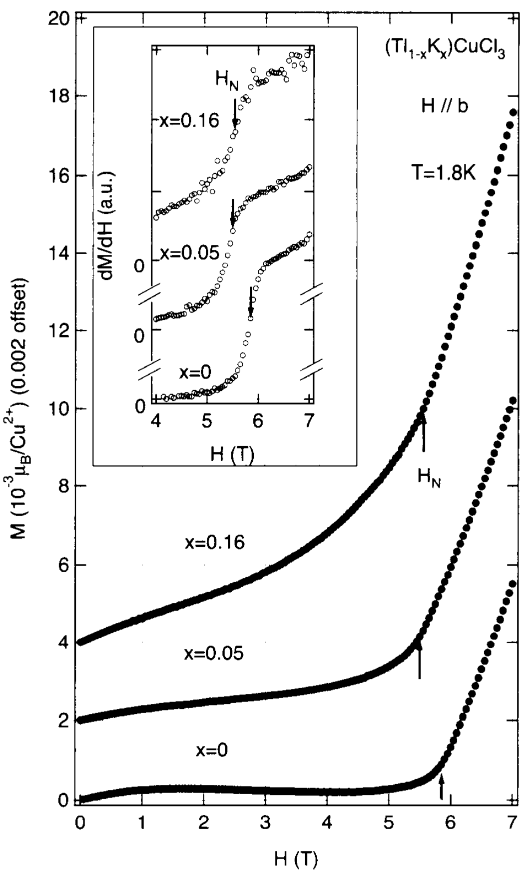

Fig. 3. The field dependence of the magnetization in $(Tl_{1-x}K_x)CuCl_3$ with $x = 0$, 0.05 and 0.16 at $T = 1.8$ K for $H \parallel b$. H_N denotes the phase transition field. The inset shows dM/dH versus H.

hysteresis was observed in the field scan. The phase transition points obtained for $T \leq 2$ K are summarized in Fig. 2.

Since the phase boundaries for $H \parallel b$ and $H \perp (1, 0, \bar{2})$ obtained by magnetization measurements coincide when normalized by the g-factor,[18] we infer that the behavior of the phase boundary is independent of external field direction. The phase boundary can be expressed by the power law $[H_N(T) - H_c] \propto T^{\phi}$, where H_c is the critical field at $T = 0$. For pure $TlCuCl_3$, the best fit is obtained with $H_c = 5.3 \pm 0.1$ T and 1.67 ± 0.07, using the data points for $H - H_c \leq 1$ T. This value of ϕ is close to $\phi_{BEC} = 3/2$ derived from the magnon BEC theory based on the Hartree-Fock (HF) approximation.[11] Recently Kawashima[20] demonstrated analytically and numerically that the critical exponent $\phi_{BEC} = 3/2$ is exact, although the HF result is usually incorrect for the critical behavior. The present result supports the BEC description of field-induced magnetic ordering in $TlCuCl_3$.

For $x \neq 0$, the exponent ϕ obtained with the data points for $H - H_c \leq 1$ T decreases systematically with increasing x, i.e., $\phi = 1.46 \pm 0.12, 1.18 \pm 0.15$ and 1.11 ± 0.15 for $x = 0.055, 0.13$ and 0.22, respectively. The value of ϕ for $x = 0.055$ becomes close to unity, when $H - H_c \leq 0.5$ T. These results indicate that randomness produces qualitative change in critical behavior. As shown in Fig. 2, the value of critical field H_c decreases with increasing x. This behavior is consistent with the x dependence of the lowest singlet-triplet excitation energy, which decreases with increasing x and reaches the bottom at about $x \sim 0.2$.[21] The excitation gap in $TlCuCl_3$ decreases under hydrostatic pressure, and $TlCuCl_3$ undergoes pressure-induced quantum phase transition to the antiferromagnetic state.[22)–24] Since the ion radius of K^+ ion is smaller than that of Tl^+ ion, substituting K^+ ions for a part of Tl^+ ions produce the compression of crystal lattice. Thus, the decrease of H_c with x should be attributed to the chemical pressure.

According to the theory by Fisher et al.,[17] the transition temperature T_c is expressed as $T_c \sim [\rho_s(0)]^x$, and $\rho_s(0) \sim (\rho - \rho_c)^\zeta$ where ρ_c is the critical density at which the superfluid transition occurs at $T = 0$, $\rho_s(0)$ is the superfluid density at $T = 0$ and exponents x and ζ are respectively $x = 3/4$ and $\zeta \geq 8/3$ for three dimensions. In the present spin system, magnetization is proportional to the density of magnon bosons ρ, and $\rho - \rho_c \sim H - H_c$ at $T = 0$. Therefore, the phase boundary of $Tl_{1-x}K_xCuCl_3$ with $x \neq 0$ is described by the power law $[H_N(T) - H_c] \propto T^\phi$ with an exponent $\phi \leq 1/2$ in the vicinity of $T = 0$. The phase boundary for $x \neq 0$ should be concave function of T and tangential to the field axis at $T = 0$. In high magnetic field region, where the density of magnons exceeds a certain value to fill up the lowest local potential, the phase transition may be described by the standard BEC transition, and the phase boundary should be expressed by the power law with the exponent $\phi \geq 3/2$. As shown in Fig. 2, the low-temperature phase boundary for $x \neq 0$ is approximately linear in temperature T. We infer that the T-linear behavior arises from the crossover from the high field region represented by the standard BEC transition to the critical region for the Bose glass-superfluid transition which is characterized by the small exponent $\phi \leq 1/2$.

Since the magnetization and the magnetic susceptibility $(\partial M/\partial H)$ corresponds to the number of magnon bosons and the compressibility of the lattice boson system, respectively, we have measured the magnetization in $Tl_{1-x}K_xCuCl_3$.[18] On the temperature scan, the magnetization exhibits a cusplike minimum at the transition temperature T_N for $H > H_c$ as predicted by the magnon BEC theory.[11] The magnetization curve at $T = 1.8$ K for $x \neq 0$ has finite slope below the critical field H_c, as shown in Fig. 3. The finite magnetization slope is not due to the finite temperature effect, because $TlCuCl_3$ exhibits almost zero magnetization up to the critical field H_c. The singlet ground state turns into the magnetic state with finite susceptibility due to exchange randomness. With increasing magnetic field, magnetization rapidly increases at a transition field H_N indicating field-induced magnetic ordering. The transition field H_N was assigned to the field with an inflection point of the magnetization field derivative, as shown in the inset of Fig. 3. The finite magnetic susceptibility below H_N for $x \neq 0$ indicates that the compressibility at $T = 0$ is finite and there is no gap. From phase boundaries shown in Fig. 2, it is apparent that there is a critical field H_c of the field-induced magnetic ordering, and that for $H < H_c$, magnetic ordering is absent in spite of the finite susceptibility. This implies that for $H < H_c$, magnons are localized due to random potential with continuous localized states. These properties are compatible with the characteristics of the Bose glass phase discussed by Fisher et al.[17] Thus, we infer that the ground state for $H < H_c$ in $Tl_{1-x}K_xCuCl_3$ is the Bose glass phase of magnons.

Figure 4 shows a schematic phase diagram for magnetic field versus interdimer interaction in $Tl_{1-x}K_xCuCl_3$. Here $\tilde{J}$ and $\bar{J}$ are expressed by a certain linear combination of interdimer interactions,[8] which determine the phase boundaries between the gapped phase and the ordered phase and between the ordered phase and saturated phase, respectively. Solid and dashed lines correspond to pure and disordered systems, respectively. Two arrows indicate the values of $|\tilde{J}|/J$ corresponding to $TlCuCl_3$ and $KCuCl_3$. The Bose glass phase is produced between gapped and ordered phases corresponding to superfluid and Mott insulating phases, respectively.[17] The field range of the Bose glass phase increases with increasing the degree of the randomness, and finally the gapped phase disappears. When the parent system is close to the quantum critical point where the gap just closes due to interdimer interactions as $TlCuCl_3$, the randomness is so effective that a small amount of randomness can wipe out the gapped phase. We consider that for this reason, the gapped phase accompanied by zero magnetization is absent in $Tl_{1-x}K_xCuCl_3$ even for $x = 0.05$.

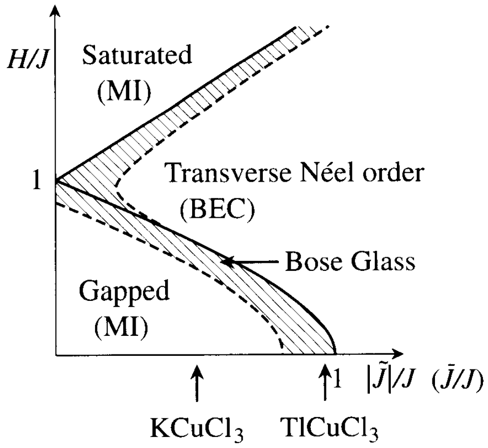

Fig. 4. Schematic phase diagram for magnetic field versus interdimer interaction in $Tl_{1-x}K_xCuCl_3$. Solid and dashed lines denote the phase boundaries for $x = 0$ and $x \neq 0$, respectively. The Bose glass phase exists in the hatched area.

§4. Conclusion

We have presented the results of specific heat and magnetization measurements performed on $Tl_{1-x}K_xCuCl_3$. Well-defined field-induced phase transition was observed. The relation between the transition field and temperature is represented by the power law $[H_N(T) - H_c] \propto T^{\phi}$. The critical exponent for pure $TlCuCl_3$ was evaluated to be $\phi = 1.67 \pm 0.07$ using the data points for $H - H_c \leq 1$ T. This value is close to $\phi_{BEC} = 3/2$ derived from the magnon BEC theory. We infer that the partial K^+ ion substitution for Tl^+ ions produce randomness in the dominant intradimer interaction, which lead to random local potential for magnon. The randomness produces a qualitative change in critical behavior. For $x \neq 0$, the phase boundary observed below 2 K is almost linear in temperature T. This behavior is interpreted as the crossover from the high field region represented by the standard BEC transition to the critical region for the Bose glass-superfluid transition. The ground state for $H < H_c$ in $Tl_{1-x}K_xCuCl_3$ with $x \neq 0$ has finite magnetic susceptibility, no gap and no long range order. These properties is consistent with those of the Bose-glass discussed by Fisher et al.[17]

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