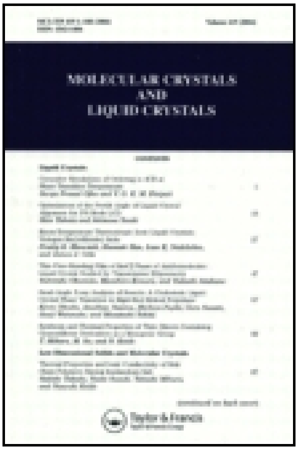

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Spin Frustration Effect in Magnetic Graphite Intercalation Compounds

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Version of record first published: 24 Sep 2006

Mol. Cryst. and Liq. Cryst., 2000, Vol. 340, pp. 1-6
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Spin Frustration Effect in Magnetic Graphite Intercalation Compounds

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The magnetic phase transition of stage-2 $Cu_cCo_{1-c}Cl_2$ GIC's has been studied using SQUID AC magnetic susceptibility. For $0.4 \leq c \leq 0.9$ the system undergoes a phase transition at T_{cl} and a reentrant spin glass transition at T_{RSG} ($<T_{cl}$). For $0.9 < c \leq 0.93$ the system undergoes a spin glass transition at T_{SG} from the paramagnetic phase to the spin glass phase. No phase transition is observed for c = 1 at least above 0.3 K. The nature of reentrant spin glass phase and spin glass phase is examined from the frequency dependence of absorption χ''.

Keywords: spin frustration effect; spin glass phase; reentrant spin glass phase; random spin system; SQUID AC susceptibility

INTRODUCTION

Random mixture graphite intercalation compounds (RMGIC's) provide model systems for studying 2D random spin systems [1-3]. Depending on combination of two kinds of ions in the intercalate layers, various kinds of spin frustration effect result from competing intraplanar exchange interactions, leading to spin glass behavior. Stage-2 $Cu_cCo_{1-c}Cl_2$ GIC's magnetically behave like a 2D XY random spin system with competing ferromagnetic and antiferromagnetic short-ranged exchange interactions. In each intercalate layer Cu^{2+} and Co^{2+} spins are randomly distributed on the triangular lattice, forming 2D random spin systems. The interaction between Co^{2+} spins is ferromagnetic, while the interaction between Cu^{2+} spins is antiferromagnetic. The interaction between Cu^{2+} and Co^{2+} spins is ferromagnetic. The sign of Θ changes around c = 0.8, indicating that the competition between intraplanar ferromagnetic and antiferromagnetic exchange interactions occurs.

In this paper we report the magnetic phase transitions of stage-2 $Cu_cCo_{1-c}Cl_2$ GIC's ($0 \le c \le 1$) using SQUID AC magnetic susceptibility with various frequencies. The nature of the reentrant spin glass (RSG) phase for $0.4 \le c \le 0.9$ and a spin glass (SG) phase is examined from the frequency (f), temperature (T), and field (H) dependence of χ''. The origin of RSG and SG phases is also discussed in terms of relevant theory.

EXPERIMENTAL PROCEDURE

The SQUID AC magnetic susceptibility of stage-2 $Cu_cCo_{1-c}Cl_2$ GIC's with c = 0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.7, 0.8, 0.88, 0.9, and 0.93 was measured using a SQUID magnetometer (Quantum Design MPMS XL-5) with ultra low field capability. First the sample was cooled in zero magnetic field. Then the SQUID AC magnetic susceptibility along the c plane was measured with increasing T from 1.9 to 15 K with and without H. Both an ac magnetic field with amplitude h (= 50 mOe) and frequency f (= 0.007 - 1000 Hz) and an external dc magnetic field H (= 0 - 1 kOe) were applied along the c plane (any direction perpendicular to the c axis).

RESULT

Figure 1 shows the T dependence of χ'' for c = 0.8 at various f. The absorption χ'' has two peaks at T_{RSG} (= 3 - 6 K) and T_{cl} (= 9.20 - 9.30 K). The peak at T_{RSG} shifts to the high temperature side with increasing f. Figure 2 shows the f dependence of χ'' for c = 0.80 at various T in the frequency range $0.007 \le f \le 1000$ Hz. Above 3.4 K χ'' has a peak, shifting to the higher frequency side with increasing T. This shift of local maximum indicates that the lowest temperature phase is a RSG phase. The broad spectral width of up to 5.7 decades in frequency FWHM (full width at half maximum) (compared to a single time Debye fixed width of 1.14 decades) reflects an extremely broad distribution of relaxation times. The maximum of χ'' vs f provides a method for determining an average relaxation time τ for each T: $\omega\tau = 1$. The inset of Fig.3 shows the T dependence of τ. The relaxation time τ dramatically increases with decreasing T in such a limited

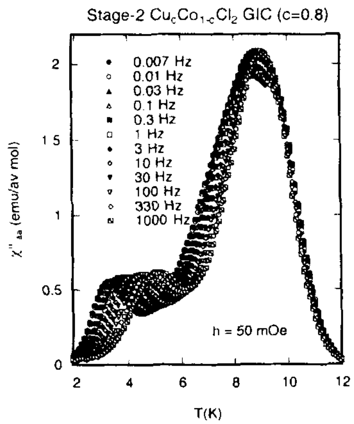

FIGURE 1 χ'' vs T for stage-2 $Cu_cCo_{1-c}Cl_2$ GIC with c = 0.8 at various f. H = 0. h = 50 mOe. h ⊥ c.

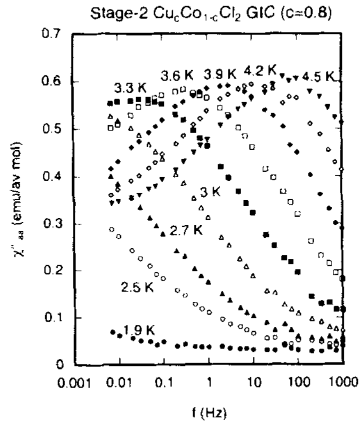

FIGURE 2 χ'' vs f for various T. c = 0.8.

temperature range. The most likely source for such a dramatic divergence of τ is a critical slowing down. The least squares fit of our data to $\tau = \tau_0(T/T^* - 1)^{-x}$ yields exponent x = 13.8 ± 1.4 and T* = 1.83 K ± 0.21 K for c = 0.80. Note that x = 8.51 for c = 0.88. Figure 3 shows a scaling plot of χ''/ω^y as a function of $\omega\tau$, where y = 0.0089 ± 0.0003. It seems that almost all the data fall on a scaling function given by

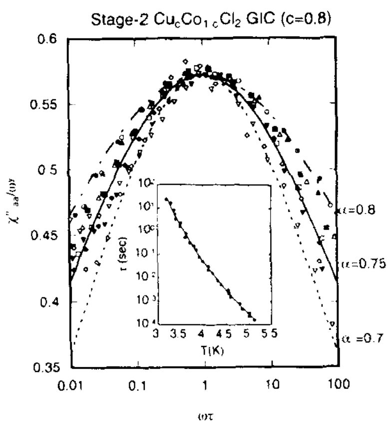

FIGURE 3 Plot of χ''/ω^y as a function of $\omega\tau$ for stage-2 $Cu_cCo_{1-c}Cl_2$ GIC with c= 0.8, where y = 0.0089 and $\tau = \tau_0(T/T^* - 1)^{-x}$ with τ_0 = 0.59 sec, x = 13.81, and T* = 1.825 K: f = 0.01 (●), 0.05 (○), 0.1 (▲), 0.5 (Δ), 1 (■), 5 (□), 10 (♦), 50 (◊), 100 (▼), and 500 Hz (∇).The scaling functions given by Eq.(1) with a = 0.7, 0.75, and 0.8 are also shown, where multiplicity constants are appropriately chosen. The inset shows a plot of τ vs T.

$$f(\omega\tau) = \mathrm{Im}[\frac{1}{1+(i\omega\tau)^{1-\alpha}}] = \frac{\cos(\pi\alpha/2)/2}{\cosh[(1-\alpha)\ln(\omega\tau)]+\sin(\pi\alpha/2)}, \quad (1)$$

with $\alpha = 0.75 \pm 0.05$ for $0.01 \leq \omega\tau \leq 100$.

For $0.9 < c \leq 0.93$ the system undergoes a spin glass transition at T_{SG} from the paramagnetic phase to the SG phase. No phase transition is observed for c = 1 at least above 0.3 K because of the fully frustrated nature of 2D antiferromagnet on the triangular lattice. Figure 4 shows the T dependence of χ'' for c = 0.93. A singe peak at T_{SG} shifts to the higher temperature side with increasing f. Figure 5 shows the f dependence of χ'' for c = 0.93. This f dependence is rather different from that for c = 0.8. The absorption χ'' decreases with increasing f below 5.9 K. Above 6.3 K it shows a peak, shifting to the higher frequency side with increasing T. Figure 6 shows the T dependence of χ'' with various H along the c plane. The peak of χ'', T_{SG}, shifts to the low temperature side with increasing H. The least squares fit of the data of T_{SG} vs H to the form $H = H_0[1-T_{SG}(H)/T_{SG}(H=0)]^a$ yields $a = 3.56 \pm 0.38$. This exponent a is much larger than that (= 1.5) predicted by Almeida-Thouless [4] for the H dependence of freezing temperature at the transition between the paramagnetic phase and the SG phase. Note that $a = 1.26 \pm 0.02$ for c = 0.8. The dramatic increase of τ with decreasing T around T_{SG} cannot be explained by a conventional critical slowing down.

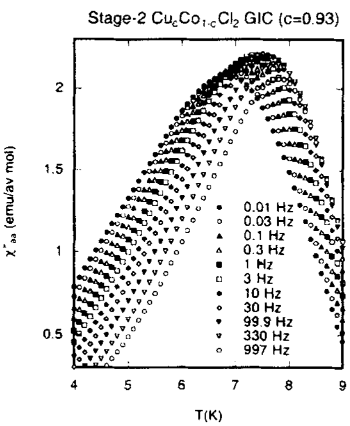

FIGURE 4 χ'' vs T for various f. c = 0.93. h = 50 mOe. h $\perp$ c.

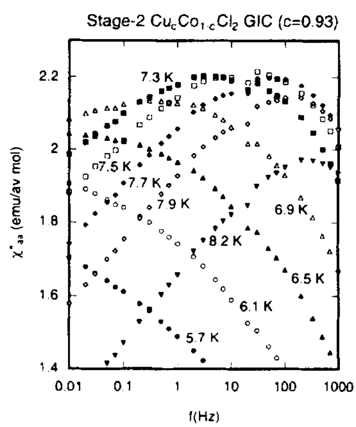

FIGURE 5 χ'' vs f for various T. c = 0.93

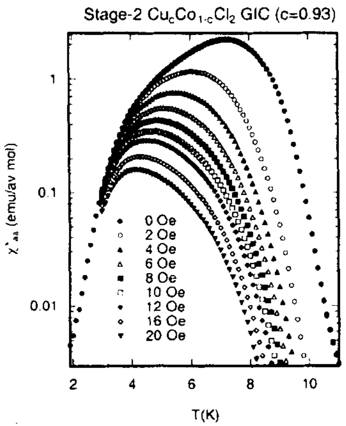

FIGURE 6 χ'' vs T for various H. c = 0.93. H ⊥ c. f = 1 Hz. h = 50 mOe.

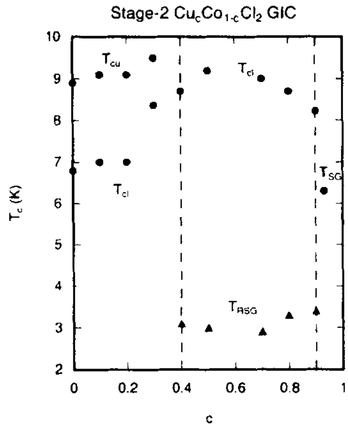

FIGURE 7 Magnetic phase diagram of stage-2 $Cu_cCo_{1-c}Cl_2$ GIC's, where peak temperatures of χ'' at f = 0.1 Hz are plotted for each Cu concentration.

These results suggests that the nature of SG phase below T_{SG} is essentially different from that of RSG phase below T_{RSG}.

DISCUSSION

Figure 7 shows the magnetic phase diagram of stage-2 $Cu_cCo_{1-c}Cl_2$ GIC's. For $0 \leq c \leq 0.3$ the system undergoes two phase transitions at T_{cu} and T_{cl} ($T_{cu} > T_{cl}$). Below T_{cu} a 2D ferromagnetic order is established in each intercalate layer. Below T_{cl} there appears a 3D antiferromagnetic phase with the 2D ferromagnetic layers being antiferromagnetically coupled along the c-axis. For $0.4 \leq c \leq 0.9$ the system undergoes a phase transition at T_{cl} and a RSG transition at T_{RSG} ($< T_{cl}$). The value of T_{RSG} is almost independent of Cu concentration. For c = 0.9 to 0.93 the system undergoes a spin glass transition at T_{SG}. For c ≈ 1 no phase transition is observed at least above 0.3 K, partly because of the frustrated nature of the 2D antiferromagnet on the triangular lattice.

Kawamura and Tanemura [5,6] have made a Monte Carlo study on the spin ordering process of the 2D ± J plane rotator (XY) model on the square lattice, where c is the concentration of AF bonds and 1-c is the concentration of F bonds. For $c \approx 0$ the system undergoes a Kosterlitz-Thouless (KT)-like transition at $T \approx J$. For $c = 0.5$ the system shows a novel type of SG transition into a chiral SG at $T \approx 0.3$ J, which is characterized with the existence of frozen-in vortices. The nature of chiral SG is not sensitive to the concentration c. For $c < c_0$ (<0.25) the reentrance phenomena are observed with the high temperature KT phase and the low temperature chiral SG phase. The Cu concentration in our system does not coincide with the concentration of antiferromagnetic bonds in the above theory, because J(Cu-Co) is ferromagnetic. The lattice form of our system is different from that in the theory. The effect of interplanar interaction on the phase transition is not also taken into account in the theory. In spite of such differences, our result is qualitatively in good agreement with the prediction from the theory: (i) T_{RSG} is almost independent of Cu concentration, and (ii) the magnetic phase diagram consists of the ferromagnetic (FM) phase for $c \leq 0.3$, the high temperature FM phase and low temperature RSG phase for $0.4 \leq c \leq 0.9$, and a SG phase for $0.9 < c \leq 0.93$.

Acknowledgments

We thank A.W. Moore and H. Suematsu for providing us with HOPG and SCKG, respectively. This work was partly supported by NSF Grant No. DMR-9201656 and 9625829.

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