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Networking and Cooperative Dynamics in Complex Physical Systems

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Abstract Phase transition and Critical phenomena in some complex physical systems are examined from the viewpoint of networking and cooperative dynamics. There are included field induced antiferromagnetism in $Cu(HCOO)_2.4H_2O$, hierarchical successive transitions in $CoCl_2$-GIC (graphite intercalation compound) and chiral glass ordering in $YBa_2Cu_4O_8$ ceramic. The characteristic features are described as the effect of staggered field, finite size and frustration in the interaction network of the systems, respectively.

1. Introduction

Phase transition and the cooperative dynamics in the natural world have long attracted a great attention of scientists. Onsager's rigorous theory showed that two-dimensional Ising (2DI) model does show a phase transition into a long range order (LRO) at a finite temperature, while 1D one does not at all[1], indicating that the connectivity of the interaction (networking) among the component atoms or molecules plays an essential role for the appearance of phase transition.

It is also shown theoretically that the critical temperature T_C decreases as cutting off the quadratic network randomly and that the phase transition disappears perfectly below the critical cutting concentration n_C[2]. Following to percolation theoretical consideration, the interaction pathway over the whole system is only one at n_C, as in the case of 1D system. It gives apparently a topological explanation of the reason why 1D system does not order at all, suggesting a basic importance of the feedback loop.

Experimentally, a phase transition in 2D lattice was first identified by proton NMR of a layer structure antiferromagnet $Mn(HCOO)_2.2H_2O$[3] and confirmed by neutron quasi-elastic scattering afterwards[4]. The value of critical exponent of the staggered susceptibility was found to be the same as that for 2DI model by precise AC magnetic susceptibility analysis[5]. Random dilution effect of 2DI model was also observed experimentally by heat capacity measurement of a dilute series, $Mn_{1-X}Mg_X(HCOO)_2.2H_2O$[6].

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In this way, it was confirmed that the feedback loop in the interaction network plays an essential role for the onset of a phase transition.

The observed critical phenomena of 2DI model is, however, surprising because the exchange interaction in the system is certainly expected of Heisenberg(H)-type from the isotropic nature of Mn^{2+} ions and 2DH model is theoretically believed not to order at all. It could be, therefore, taken as an experimental verification of an essential role of a very weak anisotropy on the onset of a phase transition and also of the universal nature of critical phenomenon.

It has been also found in the investigation process of various 2D magnets of a slightly complicated lattice structure that there are some other important factors which affect strongly on phase transition and critical phenomena. These factors are staggered field, finite size, frustration etc. in the interaction network and bring a dramatic change in the ordering characteristic from the simple system of regular and infinite lattice without frustration. In the following, such characteristic critical phenomena are reviewed and the origin of such factors are critically discussed.

2. Staggerd Field Effect on Ordering—Field Induced Antiferromagnetic Order above T_N[7]

$Cu(HCOO)_2.4H_2O$ is a 2DH like antiferromagnet of S=1/2. The magnetic susceptibility showed a broad maximum around 60 K, indicating the development of in-plane short range order (SRO)[8]. As temperature decreases further, it showed an antiferromagnetic phase transition at T_N (~ 17 K) followed by a distinct peak of magnetic susceptibility[8]. Correspondingly, NMR line shift also increases down to T_N. The magnitude of NMR line shift, however, was much larger than that estimated from the susceptibility data[9]. Besides, the NMR pattern (angular dependence of line shift) above T_N was qualitatively different from those estimated by using the susceptibility data[9], as shown in Fig. 1.

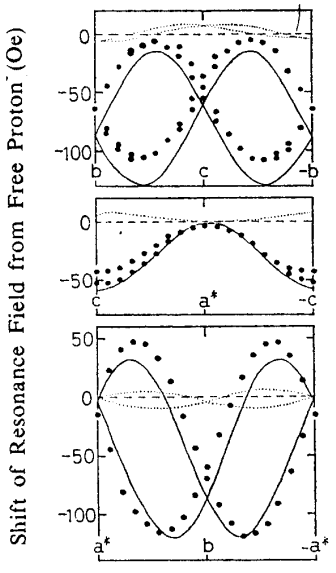


Fig.1 NMR pattern of $Cu(HCOO)_2.4D_2O$ Brack circle is experimental data. and real and dotted lines are the calculated curves with and without staggered field effect.[9]

Such anomalous phenomena can not be expected at all for a simple antiferromagnet and has not been noticed distinguishably in the ordering process of $Mn(HCOO)_2.2H_2O$ mentioned above. The present discrepancy of experimental and calculated line shift could then be excluded by assuming a large field induced staggered magnetization L above T_N

as shown also in Fig.1. The magnitude of L was found to be proportional to the intensity of external DC magnetic field H and the direction is perpendicular to the field[9,10]. Surprising was that the absolute magnitude amounted to nearly half of the saturation value at a field much weaker than the exchange field above T_N ($T \sim 1.2T_N$). The origin is reasonably explained by linear response analysis as a staggered field effect[7] as in the following.

In the compound, the local symmetry of crystalline field at two sublattice points is different to each other and gyromagnetic ratio tensor g is thus inequivarent. In such a crystallographic two sublattice (C_2) system, a staggered field H_S could be introduced effectively by applying an external (uniform) magnetic field H. As the result, a staggered magnetization L is induced in addition to the uniform magnetization M. In the paramagnetic region above T_N and at a weak magnetic field, the field induced uniform and staggered magnetizations are proportional to the field intensity H and H_S, respectively[10].

The staggered field intensity H_S is known to be of the order of $|\Delta g/g| H$, where g and Δg are the average and the difference of g-tensor components at two sublattice points and $|\Delta g/g|$ amounts to several percent in the present case. The magnitude of staggered susceptibility χ_S in antiferromagnet, which is the linear response coefficient for H_S is much larger than the uniform susceptibilityχ_U at a low temperature, especially in such a low dimensional system of S=1/2 in which the critical temperature T_N is much lower than the Curie-Weiss one. The present characteristic feature is thus attributed to both the inequivalence of g-tensors and the development of SRO in the paramagnetic region.

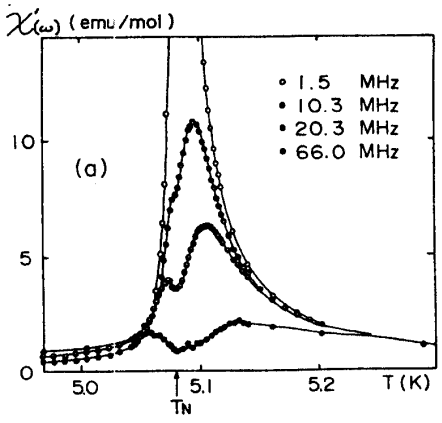
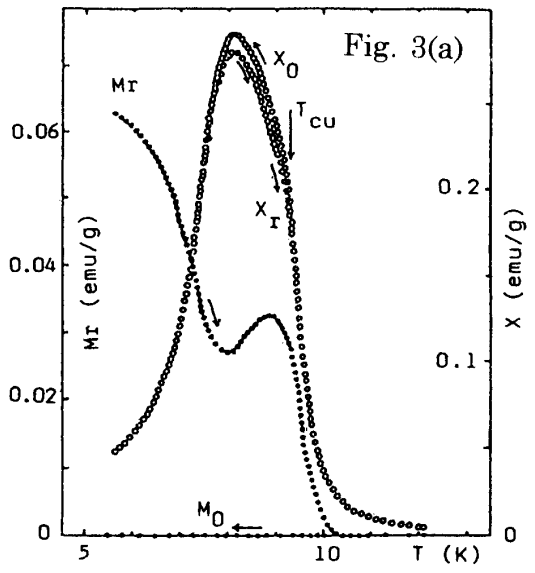


Fig. 2 $\chi'(\omega)$-T curve for various measuring frequencies. Demagnetization correction is carried out.[12]

It is remarkable here that the present antiferromagnetic order is essentially of a 2D system and free from the inter-plane interaction, because the inter-plane spin correlation is found to be ferromagnetic while the inter-plane interaction in the present case is antiferromagnetic. Such a staggered field effect was also identified in $Co(HCOO)_2.2H_2O$ by proton NMR[11]. The critical divergence and the characteristic dispersion of magnetic susceptibility at the Neel temperature (see Fig.2) was attributed to the staggered susceptibility and to the critical slowing down of staggered magnetic fluctuation, respectively[12]. For $Mn(HCOO)_2.2H_2O$, the inequivalence of g-tensor is negligibly small, which is probably the reason why such a staggered field effect could not be distinguishable.

3. Finite Size Effect on Ordering—Successive Phase Transitions of a Hierarchical Nature

Graphite compound intercalated by $CoCl_2$ ($CoCl_2$-GIC) is a layer structure ferromagnet of S=1/2[13]. Since each $CoCl_2$ layer is separated by a certain number of carbon layers (stage number), a high stage compound is thus a model of a 2D ferromagnet. Successive magnetic transitions was found in stage 2 $CoCl_2$-GIC at T_{CU} (= 9K) and at T_{CL} ($<T_{CU}$) by detailed magnetic measurement at weak field limit as shown in Fig. 3(a)[14]. Referring to the easy



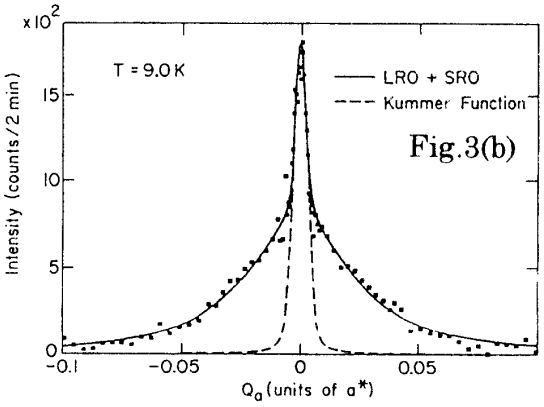


Fig. 3(a) Temperature dependence of thermoremanent magnetization Mr and linear magnetic susceptibility χ_0 of stage 2 $CoCl_2$-GIC.[17]
Fig. 3(b) Fits of structure factors based on a sum of Lorenzian and δ-function parts (solid line) and on the Kummer function (dashed line) overlaid with data at a temperature between T_{CU} and T_{CL}.[18]

plane anisotropy of six-fold symmetry and to theoretical prediction of successive transitions in such a 2DXY model[15], the intermediate state between T_{CU} and T_{CL} was first suspected to be a so called KT (Kosterlitz and Thouless) phase[16]. However, referring to a small but finite thermoremanent magnetization, a true 2D LRO not of KT-type but of conventional type was predicted in the intermediate temperature region[17]. It was then confirmed by the observation of a Bragg's ridge in neutron quasi-elastic scattering as shown in Fig.3(b)[18]. The origin of these experimental facts were reasonably explained by a hierarchical successive transitions, taking the finite size effect of each intercalated $CoCl_2$ plane into account[19], as in the following.

In a quasi 2D system, intra-plane correlation length $\xi^{(2)}$ increases divergently as temperature decreases to $T_C^{(2)}$, the critical point of ideal 2D system. Since the spins at a distance $r < \xi^{(2)}$ behaves in a coherent way, an effective enhancement of the inter-plane coupling by a factor $(\xi^{(2)}/a)^2$ takes place where a is in-plane lattice spacing. As the result, a 2D to 3D crossover of ordering happens near $T_C^{(2)}$, which results in a 3D LRO. This has been always the case for usual layer structure compounds. However, if each layer is divided into

finite size clusters of r_0, $\xi^{(2)}$ is limited geometrically. In addition, if the condition $J'/J < (a/r_0)$ is satisfied, a dimensionality crossover can not take place. Instead, the system goes into a 2D LRO below T_C [2].

Certainly this should be the case for the present stage 2 $CoCl_2$-GIC due to the good 2D nature and to the fact that each intercalated $CoCl_2$ plane is not extended infinitely and divided into finite size clusters of mesoscopic scale[14]. In such a case, 2D LRO does not immediately trigger a 3D LRO just below T_C [2], because the entropy gain by inter-plane randomness can compensate the energy gain by inter-plane interaction. Such a delay of 3D correlation necessarily introduces a hierarchy of ordering like as disorder ⇆ intra-cluster (true 2D) LRO with inter-cluster disorder ⇆ inter-cluster (3D) LRO at two successive temperatures. Such a hierarchical successive ordering phenomenon was also observed in a superconductive ceramic of $YBa_2Cu_4O_8$, in which a finite size effect is very distinguishable from the ceramic lattice structure.

4. Frustration Effect on Ordering

4-1. New Type of Ordered Phase on Chiral Degree of Freedom

Frustration is an important factor which suppresses onset of phase transition. A simple model system is triangular antiferromagnet, in which any phase transition does not appear into a spin LRO. This is the case for I model. For XY or H model, in which the interaction is isotropic, however, a new type of LRO not on spin but on chiral degree of freedom has been predicted theoretically[20,21], although any experimental verification has not been given at all, up to now. A most complicated version of frustrated system should be a spin glass. It was first proposed theoretically as a model to explain the cusp singularity which was found at the phase transition of CuMn alloy by Canella and Mydosh[22].

In the spin glass model, both ferromagnetic and antiferromagnetic interactions of I-type with various intensity are randomly distributed[23], which results in a complicated and difficult situation for ordering due to various types of frustration in addition to disappearance of feedback loops. Nevertheless, a phase transition into a glassy ordered state was predicted theoretically and named as a spin glass phase[23]. A characteristic critical phenomenon is negative divergence of nonlinear magnetic susceptibility χ_2[24], which was actually observed in a AuFe alloy by AC magnetic measurements[25]. For the spin glass model of H- or XY-type interaction, on the other hand, a phase transition was suggested not to take place but into a new glassy state on chiral degree of freedom i.e. a chiral glass phase[26,27]. It is characterized by a negative divergence of nonlinear susceptibility on a conjugate field to chirality

4-2. Chiral Glass Ordering in Superconductive Ceramics

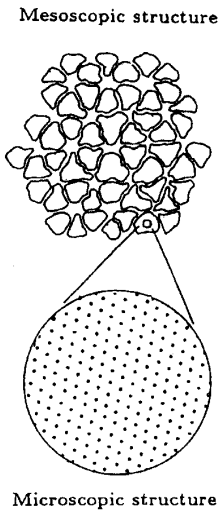


Fig. 4 Schematic view of ceramic structure

Generally a ceramic is composed of crystalline mesoscopic clusters which are coupled mutually through random interface boundaries, as shown schematically in Fig.4. A ceramic is thus regular in microscopic scale but random in mesoscopic scale. The ordering of the system will necessarily proceed successively from the intra-cluster to the inter-cluster direction in a hierarchical way as already discussed in the previous paragraph.

$YBa_2Cu_4O_8$ is one of so called high-Tc superconductors. Different from other high-Tc materials, it is stoichiometric and free from microscopic randomness due to oxygen vacancy. It is also free from twin formation and prepared in a single superconductive phase. $YBa_2Cu_4O_8$ is thus perfectly regular crystallographically and the well sintered specimen should be a typical example of superconductive ceramics. Successive superconductive transitions have been observed in such a specimen at T_{C1} (= 80 K) and at T_{C2} ($<T_{C1}$).[28] The ordering characteristic is shown in Fig.5.

In the temperature region between T_{C1} and T_{C2}, thermoremanent magnetization Mr perfectly disappears and field cooled and zero-field cooled magnetization, M_{FC} and M_{ZFC} agree with each other. Since these are found to be proportional to the external field intensity, the diamagnetism is well described due to Meissner effect inside each grain. Below T_{C2}, on the other hand, the magnitude of M_{ZFC} is much larger than M_{FC}. The diamagnetism is then attributable to flux shielding by intergrain superconducting network. Correspondingly, electric resistance was found to decrease in a two-step way and almost disappear below T_{C2}. The successive superconductive ordering is thus identified to be a hierarchical one from the intragrain to inter-grain direction.

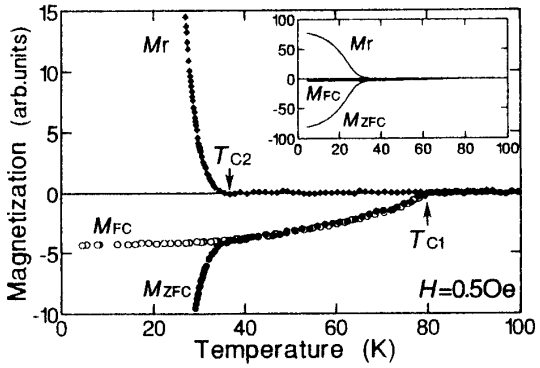


Fig. 5 Temperature dependence of field cooled, zere field cooled and thermoremanent magnetizations, M_{FC}, M_{ZFC} and Mr of $YBa_2Cu_4O_8$ ceramic.[28]

The inter-grain interaction in the present system is of a Josephson-type and can be expressed in terms of exchange interaction of XY-type. The coupling constant is always positive for a conventional metal superconductor. It could be either positive or negative for

an anisotropic superconductor like $YBa_2Cu_4O_8$, which introduces frustration here and there in the Josephson-coupled interaction network. The inter-grain ordering is thus described by that of 3D XY spin glass. Therefore, a phase transition into a chiral glass phase is expected at T_{C2} of the present system. Since a weak magnetic field in a Josephson coupled network is found to be a conjugate field to chirality along each placket on the network, a chiral glass ordering could thus be distinguished by a negative divergence of χ_2.

From the frequency and amplitude dependence of a nonlinear magnetic response around T_{C2}, a negative divergence of χ_2 was concluded at T_{C2}.[29] An experimental indication is given in Fig.6.[29] In qualitative agreement with theoretical prediction[30], it indicates an onset of chiral glass ordering at T_{C2}. Such an anomaly could not be found so far around T_{C1}.

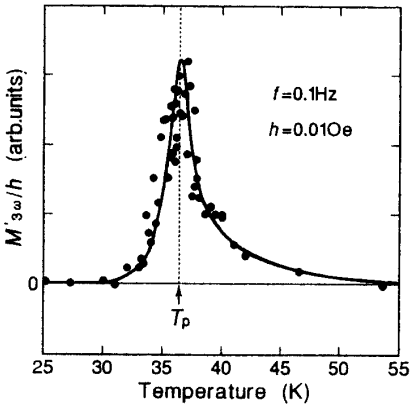


Fig. 6 Temperature dependence of 3rd harmonic component of AC response around T_{C2}[29] The value of T_P in the figure agrees with T_{C2} of this sample.

Theoretically, linear electric resistivity ρ_0 was predicted not to be exactly zero at and below the chiral glass ordering temperature, although the magnitude was actually very small.[31] The linear and nonlinear resistivity ρ_0 and ρ_2 around T_{C2} were thus examined in detail by AC electric response observation. The value of ρ_0 was concluded not to be zero around T_{C2}, while ρ_2 was found to show a divergence at T_{C2}.[32] Combined with the negative divergence of χ_2, the chiral glass ordering is thus confirmed to be realized in the inter-grain ordering of a $YBa_2Cu_4O_8$ ceramic.

5. Summary

In this paper, the ordering characteristics in some complex physical systems are examined from the viewpoint of networking and cooperative dynamics. Many such important factors in the cooperative dynamics as feedback loop, local inequivalence, finite size, randomness, frustration etc. are considered to exist also in general systems including biological, engineering or social systems and to affect on the ordering in some analogous ways. Application of the present research to such general non-physical systems may, therefore, be interesting and helpful for optimization of the systems or realization of some ideal community.

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