

## Appearance of the spin-Peierls phase under pressure in $\text{Cu}_{1-x}\text{Mg}_x\text{GeO}_3$

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The effect of pressure is studied on highly Mg-doped  $\text{CuGeO}_3$ , which at ambient pressure shows an antiferromagnetic transition but no spin-Peierls transition. The magnetic susceptibility under pressure was measured; the broad maximum around 60 K shifts to lower temperatures and the spin-Peierls (SP) transition reappears. The temperature-concentration phase diagram at various pressures was constructed and it was found that the SP phase region enlarges with pressure. These results mean that the enhancement of frustration due to nearest-neighbor and next-nearest-neighbor exchange interactions overcomes the effect of interchain interaction, and that the spin-Peierls phase is more stabilized than the paramagnetic or antiferromagnetic phases. In the temperature-pressure-concentration phase diagram, the compositional phase transition between the dimerized-antiferromagnetic phase and the uniform-antiferromagnetic phase is second order at  $P > 0.2$  GPa, but first order at  $P < 0.2$  GPa. We propose that the change in order of this transition is also due to enhancement of the frustration. We show an example of frustration effects in a one-dimensional quantum spin system.

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### I. INTRODUCTION

$\text{CuGeO}_3$  (Ref. 1) is one of the most interesting materials for studying quantum spin systems. Unlike many other spin-Peierls (SP) materials it is an inorganic cuprate allowing the growth of high-quality single crystals and impurity-doped crystals. This material is well known as a spin-Peierls material, but it has further features. One is a relatively large interchain interaction compared to the intrachain interaction [ $J'/J \sim 0.1$  (Ref. 2)], and the system appears to lie near the phase boundary between the SP and Néel phases in the phase diagram of spin-phonon coupling vs interchain interaction.<sup>3</sup> This may be why an exotic antiferromagnetic (AF) phase is observed in  $\text{Cu}_{1-x}\text{M}_x\text{GeO}_3$  ( $M = \text{Zn}, \text{Ni}, \text{Mn}$ ) and  $\text{CuGe}_{1-y}\text{Si}_y\text{O}_3$ .<sup>4,5</sup> The novelty of this AF phase is that the SP and AF order parameters coexist below the AF transition temperature, a fact confirmed by inelastic neutron scattering.<sup>6,7</sup> The ground state of the impurity-doped SP system has been studied theoretically, and the envelopes of the staggered moment and the lattice displacement of dimerization can be expressed by elliptic functions; they are in long-range order (LRO).<sup>8</sup> This model is consistent with  $\mu\text{SR}$  experiments.<sup>9</sup>

By producing many high-quality single crystals and performing composition analysis, Masuda *et al.* recently found that  $\text{Mg}^{2+}$  is the best nonmagnetic impurity for homogeneous impurity-doped  $\text{CuGeO}_3$ . They obtained a temperature vs concentration phase diagram<sup>10-13</sup> and found that the SP phase is suppressed drastically by impurity doping and that SP LRO disappears at a critical concentration ( $x_c$ ). At  $x_c$  a discontinuity was observed in AF temperature, suggesting a compositional first-order phase transition between the exotic AF phase—the dimerized-antiferromagnetic (DAF) phase, which has both the SP and AF order parameters—and a con-

ventional AF phase, the uniform-antiferromagnetic (UAF) phase. A detailed study reveals that a separation of the DAF and UAF phases exists around  $x_c$ ;  $0.027 \gtrsim x \gtrsim 0.023$ .<sup>11,13,14</sup>

Another unique feature is that this material exhibits the frustration of the interactions among spins. The angle of the Cu-O2-Cu bond is about  $100^\circ$ ,<sup>15</sup> which is close to  $90^\circ$ , and it is expected that the next-nearest-neighbor (NNN) interaction ( $J_{\text{NNN}}$ ) cannot be neglected. For simplicity we neglect the interchain interaction here. If the NNN interaction is antiferromagnetic it competes with the nearest-neighbor (NN) interaction ( $J_{\text{NN}}$ ), and frustration of interactions exists. Frustration in  $\text{CuGeO}_3$  was proposed to explain the discrepancy in the magnetic susceptibility between experiment and the theoretical Bonner-Fisher curve in the high-temperature region.<sup>16,17</sup> The Hamiltonian of  $\text{CuGeO}_3$  above the spin-Peierls transition temperature ( $T_{\text{SP}}$ ), is therefore approximated as

$$H = J \sum_i (\mathbf{S}_i \cdot \mathbf{S}_{i+1} + \alpha \mathbf{S}_i \cdot \mathbf{S}_{i+2}), \quad (1)$$

by considering only intrachain interactions. Here  $\alpha = J_{\text{NNN}}/J_{\text{NN}}$ , and  $\alpha$  is reported to be 0.24 (Ref. 16)  $\sim$  0.36 (Ref. 17). It is well known that a frustrated spin system has a finite spin gap above a critical value of  $\alpha$  ( $\alpha_c$ ).<sup>18</sup> For a one-dimensional (1D) system  $\alpha_c$  is reported to be 0.24  $\sim$  0.30 (Refs. 19–21), and in  $\text{CuGeO}_3$   $\alpha$  is quite close to  $\alpha_c$ . The reason that  $\alpha$  for  $\text{CuGeO}_3$  seems to exceed  $\alpha_c$  is that Eq. (1) neglects the interchain interaction. The values of  $\alpha_c$  given in Refs. 19–21 are for an ideal 1D system. To claim just the existence of rather large  $\alpha$ , however, the simple model Hamiltonian Eq. (1) would not be inappropriate as Refs. 16 and 17 discussed. Some experimental results suggest that  $J$  is suppressed and that  $\alpha$  is enhanced under hydrostatic

TABLE I. Samples of  $\text{Cu}_{1-x}\text{Mg}_x\text{GeO}_3$ .

Sample	$x$	
A	0.0	$x < x_c$
B	0.020 ( $\pm 0.0002$ )	$x < x_c$
C	0.026 ( $\pm 0.0002$ )	$x \sim x_c$
D	0.032 ( $\pm 0.0003$ )	$x > x_c$
E	0.035 ( $\pm 0.0003$ )	$x > x_c$
F	0.040 ( $\pm 0.0004$ )	$x > x_c$
G	0.045 ( $\pm 0.0004$ )	$x > x_c$

pressure.<sup>22,23</sup> The SP transition temperature is reported to increase with the pressure ( $dT_{\text{SP}}/dP = 4.8$  K/GPa).<sup>24</sup> References 22 and 23 therefore claim that an increase in  $\alpha$  stabilizes the SP ground state, and a numerical study shows that  $T_{\text{SP}}$  increases with increasing  $\alpha$ .<sup>25</sup>

The ground state of highly doped  $\text{CuGeO}_3$  is the Néel state, but it should be close to the phase boundary between the SP and the Néel phases. We may control  $\alpha$  by pressure, and the SP phase should appear even in highly doped  $\text{CuGeO}_3$ . The present paper studies the effect of pressure in Mg-doped  $\text{CuGeO}_3$ . We observe the reappearance of the SP phase and obtain a temperature-pressure-concentration ( $T$ - $P$ - $x$ ) phase diagram from the magnetic-susceptibility measurements. The reappearance of the SP phase is verified by the synchrotron x-ray diffraction. The phase diagram shows that the compositional phase transition above a critical pressure is a second-order transition, but first order at ambient pressure.

## II. EXPERIMENTAL DETAILS

Single crystals were prepared by the floating-zone method. Composition analysis was performed by inductively coupled plasma atomic emission spectroscopy. The compositions of  $\text{Cu}_{1-x}\text{Mg}_x\text{GeO}_3$  studied are  $x=0$  (sample A), 0.020 (sample B), 0.026 (sample C), 0.032 (sample D), 0.035 (sample E), 0.040 (sample F), and 0.045 (sample G). Sample C is on the compositional phase boundary ( $x_c$ ) in Ref. 11, and shows the SP transition and the double AF transitions. Samples D–G are above  $x_c$  and show no SP transition in magnetic susceptibility measurements (see Table I). Magnetic susceptibility was measured using a commercial superconducting quantum interference device (SQUID) magnetometer ( $\chi$ -Mag, Conductus, Ltd.). All measurements were performed with the magnetic field along the crystallographic  $c$  axis; the field strength was 1000 Oe. We used a hydrostatic high-pressure microcell developed by Uwatoko *et al.*<sup>26</sup> in a commercial SQUID magnetometer. The pressure at low temperature (LT) was calibrated by measuring the superconducting transition temperature of Pb. The relation between the pressures at LT and at room temperature (RT) is reproducible ( $P_{\text{LT}} \sim P_{\text{RT}} - 0.2$  [GPa]). From susceptibility measurements of undoped  $\text{CuGeO}_3$  we determined the pressure dependence of the SP transition temperature; it increases linearly with pressure, a result consistent with previous experiment.<sup>24</sup> This assures us that our pressure calibration at LT works well. The

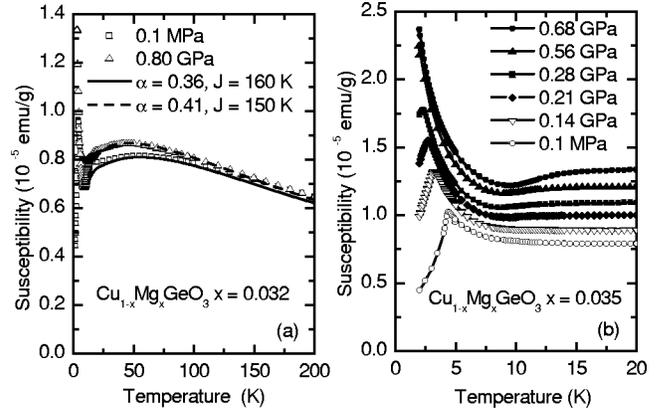


FIG. 1. (a) Magnetic susceptibility of sample D at  $2 \text{ K} < T < 200 \text{ K}$ . Solid lines show the susceptibility calculated from Eq. (1). (b) Detailed behavior of the magnetic susceptibility at differing pressures in the low-temperature region for sample E. A decrease due to SP transition is clearly observed at  $P \geq 0.021$  GPa. Data are shifted vertically for clarity.

cell is made mainly of CuBe alloy, and is slightly magnetized in the magnetic field. We therefore tested several  $\text{CuGeO}_3$ 's of differing volume and found that 20 mg is necessary for the measurement, implying that about  $2 \times 10^{-4}$  emu is the lower limit for the pressure cell. X-ray-diffraction measurements under hydrostatic pressure were performed at facilities BL-1B and 4C of the Photon Factory (KEK, Tsukuba). The wavelength was tuned to  $0.7 \text{ \AA}$  by a Si (111) double-crystal monochromator. A cylindrically shaped imaging plate was used as a detector, and a diamond anvil cell was used as a pressure cell. We calibrated the pressure at LT by measuring the lattice constant of NaCl powder.

## III. EXPERIMENTAL RESULTS

Figure 1(a) shows the magnetic susceptibility  $\chi(T, P)$  of sample D ( $x=0.032$ ).  $\chi(T, 0.1 \text{ MPa})$  has a broad maximum at 60 K; the behavior at high temperature is qualitatively the same as that of undoped  $\text{CuGeO}_3$ . Application of hydrostatic pressure shifts the maximum in  $\chi(T, P)$  to lower temperatures. The broad maximum of the magnetic susceptibility arises from the development of AF short-range order, so that this shift implies the suppression of intrachain interaction or greater frustration of the interactions among spins.

The solid lines show the magnetic susceptibility calculated from Eq. (1) when  $\alpha=0.36$  and  $J=160$  K for 0.1 MPa and when  $\alpha=0.41$  and  $J=150$  K for 0.80 GPa. The fitting parameters are determined such that the absolute values of the broad maximum of the susceptibility and of the temperature agree with the observations. The calculation is described in detail in Ref. 27. The temperature intervals in the susceptibility measurement are 5 K, and the interval of  $\alpha$  in the fitting is 0.01. The accuracies of the fitting parameters are therefore  $\Delta J = 5$  K and  $\Delta \alpha = 0.01$ . The fit of the data to the one-dimensional model [Eq. (1)] is good, and we conclude that application of hydrostatic pressure suppresses the intrachain interaction and enhances the frustration between NN and NNN spin interactions in highly doped  $\text{CuGeO}_3$ , which

shows no SP transition at ambient pressure.

The parameter  $\alpha$  seems to exceed the critical value  $\alpha_c$  for the appearance of the spin gap without dimerization.<sup>19–21</sup> The reason is that we omitted the interchain interaction in the calculation.<sup>17</sup> Therefore, the absolute value of  $\alpha$  in our calculation itself is not of much importance but we can safely conclude that the increase of  $\alpha$  with the applied pressure is genuine.

According to Fig. 2 of Ref. 28, the spin-phonon coupling term suppresses the magnetic susceptibility over the whole temperature range. The magnetic susceptibility is independent of the hydrostatic pressure at  $T \geq 130$  K. We can therefore safely assume that the spin-phonon coupling is independent of the hydrostatic pressure.

$\chi(T, 0.80$  GPa) shows a sudden drop around 10 K, but no anomaly is observed in  $\chi(T, 0.1$  MPa). To study this feature we measured the magnetic susceptibility under pressure in more detail. Figure 1(b) shows  $\chi(T, P)$  of sample E ( $x = 0.035$ ) at low temperatures. The zero of the ordinate is for  $\chi(T, 0.1$  MPa), and other data are shifted every  $+10^{-6}$  emu/g. For  $\chi(T, 0.1$  MPa) a sharp peak is observed at 4.5 K due to the AF transition; the transition temperature ( $T_N$ ) decreases monotonically with the pressure. The suppression of the AF phase is consistent with the result of the fitting of the susceptibility at high temperature seen in Fig. 1(a), since the decrease in  $J$  and the increase in  $\alpha$  both suppress the AF long-range order. Above  $T_N$ , no anomaly is observed at  $P \leq 0.14$  GPa but a slight decrease begins to be visible at  $P \sim 0.21$  GPa. The magnitude of the decrease, and the temperature, at which the susceptibility begins to decrease, increase with pressure at  $P \geq 0.21$  GPa.

The  $T_{SP}$  value of lightly doped  $\text{CuGeO}_3$  is about 10 K, and the pressure-induced anomaly in sample D or E apparently represents the SP transition. We, however, have to be careful in concluding this. In this material  $\alpha$  is close to the critical value, and we observe enhancement in  $\alpha$  in Fig. 1(b). If  $\alpha$  depends on temperature and goes beyond the critical value with decreasing temperature, the spin gap without lattice dimerization (pointed out by Haldane<sup>18</sup>) would occur at low temperature. To settle the issue we performed the synchrotron x-ray diffraction on sample E at  $P = 0.1$  MPa and 1.0 GPa.

In the inset of Fig. 2 we observe  $(1/2, 1, 3/2)$  superlattice reflection at  $P = 1.0$  GPa, which corresponds to SP dimerization<sup>29</sup> and confirms the absence of the superlattice peak at 0.1 MPa. The ratio of the superlattice peak intensity at 9.0 K and  $P = 1.0$  GPa to that of undoped  $\text{CuGeO}_3$  at the same temperature and ambient pressure was determined as 0.15 through intensity calibration based on the fundamental reflections. The temperature dependence of the integrated peak intensity is shown in Fig. 2. The peak intensity increases as the temperature decreases, and we obtained  $T_{SP} \sim 15.2$  K, which is consistent with the magnetic-susceptibility measurements. This proves that pressure genuinely causes the SP transition in sample E.

We measured  $\chi(T, P)$  for all samples and obtained the  $T$ - $P$  phase diagrams for samples B–G, as shown in Fig. 3. Monotonic increase of  $T_{SP}$ 's is observed in all Mg-doped

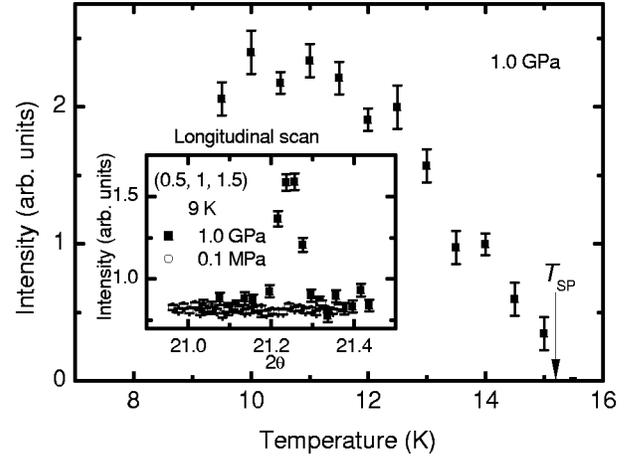


FIG. 2. Superlattice peak intensity at  $(1/2, 1, 3/2)$  in sample E at  $P = 1.0$  GPa. The inset is the peak profile of a longitudinal scan at 0.1 MPa (circles) and 1.0 GPa (squares).

samples. In samples D–G the SP phase appears above a critical pressure ( $P_c$ ). The vertical dotted lines in Fig. 3 show  $P_c$ . In all samples  $T_N$  decreases monotonically with increasing pressure. The concentration of sample C is the critical

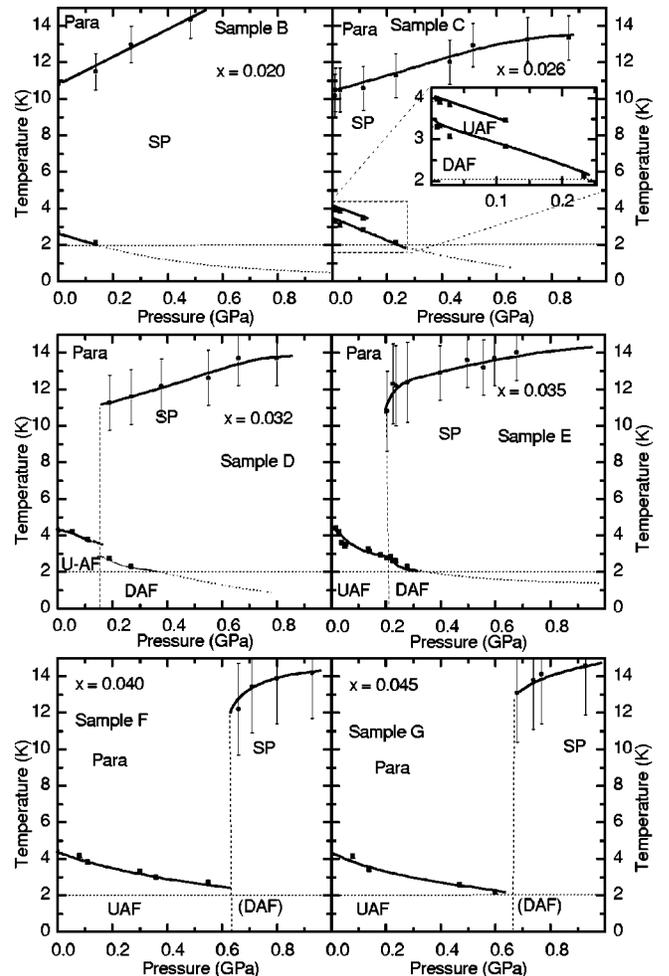


FIG. 3. The  $T$ - $P$  phase diagram in samples B–G.

concentration for the compositional phase transition between the previously reported DAF and the UAF phases.<sup>10</sup> Some of us have reported the double peaks in  $\chi(T)$  at AF transitions,<sup>11</sup> and they proposed that the phase separation of the DAF and the UAF phases exists and that the phase transition between the DAF and the UAF phases is first order.<sup>10,11</sup> In sample C, the higher-temperature transition disappears at about 0.2 GPa and the two peaks do not merge. In sample D the discontinuity is seen at  $P_c$ . On the other hand,  $T_N$  decreases monotonically in sample E over the whole pressure range, and no discontinuity is observed at  $P_c$ . This means that the compositional phase transition between the DAF and UAF phases at  $x \geq 0.035$  is a second-order phase transition.

Figure 4 shows  $T$ - $x$  phase diagrams at various pressures. The vertical dotted lines indicate the phase boundary at the critical concentration  $x_c$ . The region of the SP phase enlarges with pressure;  $T_{SP}$  increases,  $T_N$  decreases, and the phase boundary between DAF and UAF phases increases with pressure. This means that pressure stabilizes the SP phase more than the paramagnetic or antiferromagnetic phase in  $\text{Cu}_{1-x}\text{Mg}_x\text{GeO}_3$ .

At ambient pressure, two  $T_N$ 's are observed in sample C ( $x \sim x_c$ ). This is due to the phase separation between the DAF and the UAF phases; the compositional phase boundary between the DAF and the UAF phases corresponds to this sample, as reported previously.<sup>10,11,13,14</sup> At ambient pressure two  $T_N$ 's are plotted at  $x \sim x_c$  due to phase separation between the DAF and the UAF phases. At 0.2 GPa, on the other hand, there is no coexistence of  $T_{N1}$  and  $T_{N2}$ , but a sudden increase in  $T_N$  is apparent between  $x=0.035$  and 0.040.

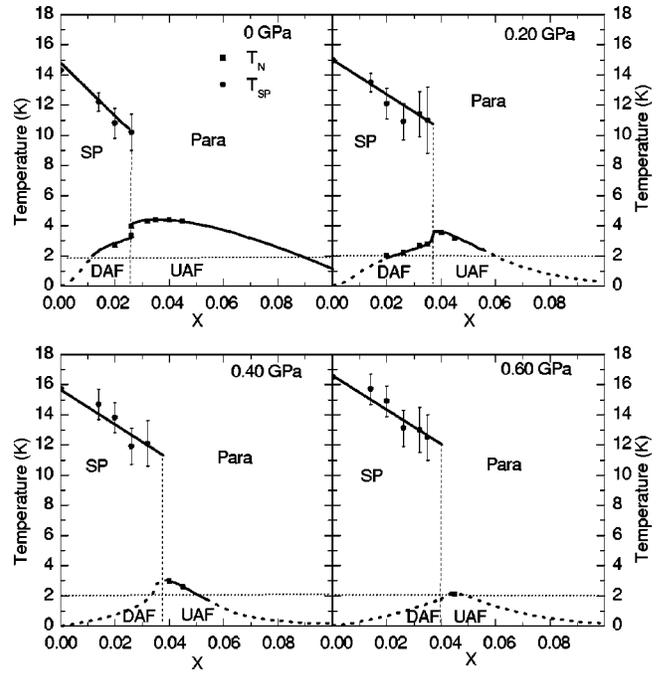


FIG. 4.  $T$ - $x$  phase diagrams under the pressure. Solid and broken lines are the guides to the eyes. Vertical dotted lines denote the phase boundary at the critical concentration  $x_c$ . Horizontal dotted lines show the low-temperature limit of the SQUID magnetometer.

The change in  $T_N$  at  $x \sim x_c$  should be easier to see in Fig. 3. Discontinuity of  $T_N$  is observed in the  $T$ - $P$  phase diagram in samples C and D but not in sample E. The schematic  $T$ - $P$ - $x$  phase diagram is therefore as in Fig. 5. The composi-

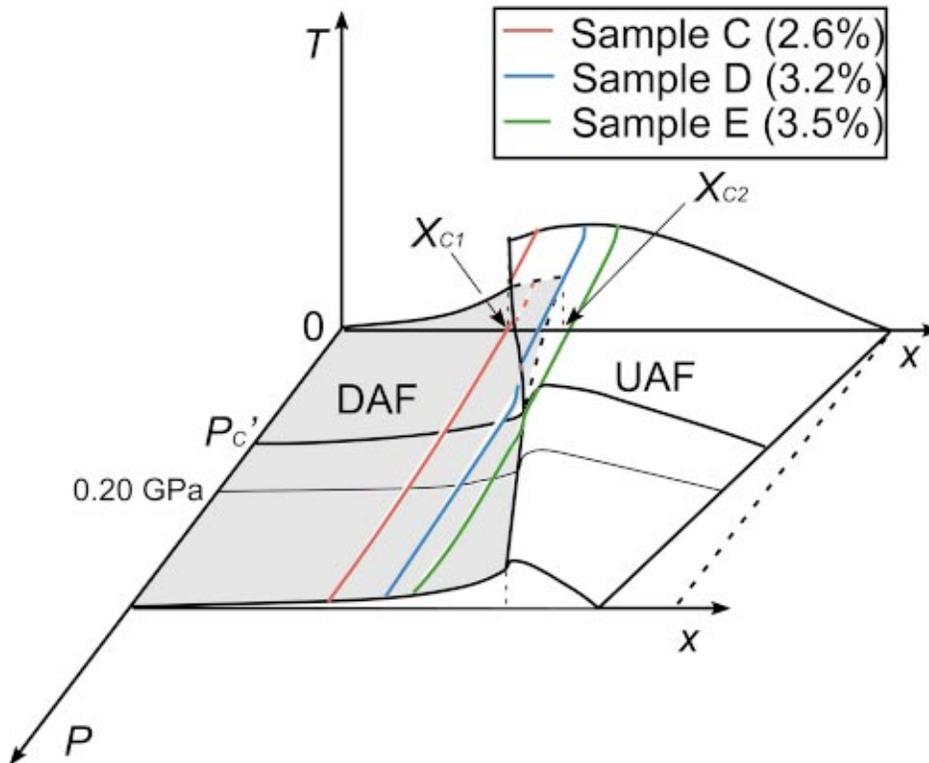


FIG. 5. (Color) The schematic  $T$ - $P$ - $x$  phase diagram.

tional phase transition between the DAF and the UAF phases is first order at low pressure, and we propose that it becomes second order above a critical pressure ( $P'_c$ ).

#### IV. DISCUSSION

We now discuss why pressure stabilizes the SP phase. The parameters that determine the phase in this system are the nearest-neighbor interaction  $J$ , the interchain interaction  $J'$ , and the frustration factor  $\alpha$  in Eq. (1). Spin-phonon coupling is also important in the SP system, but we assume that it is independent of pressure and that its effect is negligibly small. We found that the pressure suppresses  $J$  and enhances  $\alpha$  in Eq. (1), based on the magnetic-susceptibility measurements in the high-temperature region (Fig. 1). Nishi *et al.* recently performed inelastic neutron scattering with pure  $\text{CuGeO}_3$  at ambient pressure and 2 GPa.<sup>30</sup> Dispersion of the spin excitation along the  $c^*$  axis (the 1D-chain axis) was suppressed, and that along the  $b^*$  axis (perpendicular to the 1D-chain axis) was unchanged by the pressure. This means that the effective interchain interaction,  $J'/J$ , is enhanced by the pressure. Conventional SP theories of a pure system<sup>3</sup> and doped system<sup>31</sup> which do not consider the frustration term, indicate that enhancement of the interchain interaction stabilizes the Néel phase, rather than the SP phase. The experimental result, however, is different: the SP phase is more stabilized and even reappears in highly doped samples as a result of pressure. Hence, apparently the reappearance of the SP phase is not due to enhancement of the interchain interaction by the pressure and we must take account of the frustration factor  $\alpha$ . From the density-matrix–renormalization-group study on undoped  $\text{CuGeO}_3$  one can see that the SP phase is stabilized with increasing  $\alpha$  in Fig. 3 of Ref. 25. We therefore suppose that  $J'$  and  $\alpha$  compete with each other in this system. At ambient pressure in highly doped  $\text{CuGeO}_3$ ,  $J'$  overcomes the effect of  $\alpha$  and the UAF phase appears. Application of pressure enhances  $\alpha$  and the SP, and the DAF phases appear.

The compositional phase transition between the DAF and

the UAF phases is first order at low pressure. We propose that it becomes second order at 0.2 GPa, as shown in Fig. 5; the phase boundary between these two phases changes at a critical pressure,  $P'_c$ , from first order to second order as the pressure increases. Saito studied the compositional phase transition at  $\alpha=0$  using the phase Hamiltonian method,<sup>31</sup> and asserted that the phase transition is first order when  $J'$  is large and second order when  $J'$  is small. The experimental result shows, however, that the phase transition changes from first order to second order with  $J'$ , since neutron scattering indicates that  $J'$  increases with pressure.<sup>30</sup> This fact also tells us that  $\alpha$  must be considered in order to explain the phase diagram. We conclude that  $J'$  and  $\alpha$  compete, and  $\alpha$  dominates the effect of  $J'$  at higher pressures. The competition between  $\alpha$  and  $J'$  plays an important role in this system.

#### V. SUMMARY

In summary, the application of hydrostatic pressure causes the SP phase to reappear in highly doped  $\text{CuGeO}_3$ , which shows no SP transition at ambient pressure. The phase of this system is determined by competition between the interchain interactions and frustration between NN and NNN interactions. We have obtained the temperature-pressure-concentration phase diagram and verified that frustration plays an important role in these features.

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