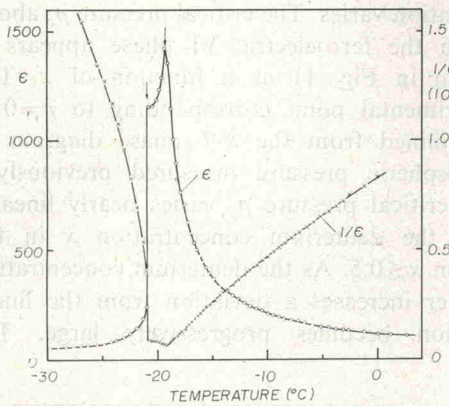

Fig. 1. Temperature dependence of the dielectric constant ε (○) and its reciprocal $1/\varepsilon$ (×) along the c^*-direction of $\{(NH_4)_3H(SO_4)_2\}_{0.03}\{(ND_4)_3D(SO_4)_2\}_{0.97}$ at $p=3.72$ kbar. Vertical arrows indicate the dielectric anomalies at the II-IX and IX-VI transitions. Frequency: 100 kHz.

$1/\varepsilon$ along the c^*-direction of the deuterated compound at $p=3.72$ kbar. As shown by the arrows, two anomalies are seen around the II→VI transition region. These anomalies indicate that there is an interdediate phase in a narrow temperature region between the room temperature phase of II and the ferroelectric VI phase. The intermediate phase is denoted as Phase IX. Figure 2 indicates the temperature dependence of the inverse of the dielectric constant $1/\varepsilon$ at pressures higher than 6 kbar. A clear break is seen for each curve as indicated

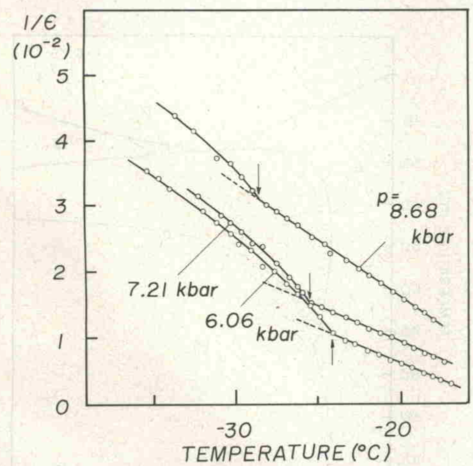

Fig. 2. Temperature dependence of the reciprocal of the dielectric constant $1/\varepsilon$ along the c^*-direction of $\{(NH_4)_3H(SO_4)_2\}_{0.03}\{(ND_4)_3D(SO_4)_2\}_{0.97}$ at different hydrostatic pressures. Vertical arrows indicate the anomalies at the VIII-VI transition. Frequency: 100 kHz.

by an arrow. The dielectric anomaly corresponds to a phase transition from the room temperature phase of II to a new intermediate phase which is denoted as Phase VIII. The pressure-induced phases of VIII and IX were not noticed in our preliminary work.[6)]

From the results of dielectric constant measurements, we can obtain the pressure-temperature phase diagrams for different concentration x. Figures 3∼8 show the p-T phase diagrams for compounds with $x=0$, 0.14, 0.40, 0.60, 0.79, and 0.97, respectively. The phase diagram of the normal compound shown in Fig. 3 was the one reported previously.[3)] As the deuterium concentration increases the two pressure-induced ferroelectric phases VI and VII appear in lower pressure region. The intermediate phase IX can be seen in the pressure region studied for the compounds with $x\gtrsim 0.60$.

In the normal compound the II-III phase boundary was not represented by a linear relation, but it was approximated by a quadratic form of $\Theta_{\text{II-III}}=T^0_{\text{II-III}}+Kp+\gamma p^2$.[9)] We estimated the parameters $T^0_{\text{II-III}}$, K, and γ as fimctions of x from the phase diagrams. The results are shown in Fig. 9. The II-III transition temperature $T^0_{\text{II-III}}$ at 0 kbar and the initial pressure slope K vary with concentration x very slightly. On the other hand, the parameter

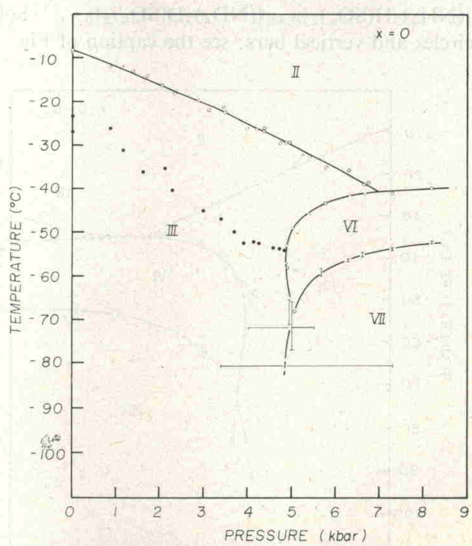

Fig. 3. Pressure-temperature phase diagram of $(NH_4)_3H(SO_4)_2$.[3)] Solid circles show the position of the broad dielectric constant peak at constant-pressure runs. Short bars indicate temperature (or pressure) hysteresis of the first order transitions.

γ seems to change its sign as x varies from 0 to 0.97.

Figure 10 shows the pressure dependence of the inverse of the maximum value of the dielectric constant at the diffuse peak in Phase III for different x. The relation between $1/\varepsilon_{max}$ and pressure p is linear for each compound, that is, a Curie-Weiss like relation $1/\varepsilon_{max}=C^*(p_0-p)$ is held. The relations between $1/\varepsilon_{max}$ against p for various deuterium concentrations x are almost parallel. Therefore the constant C^* is practically unchanged as deuterium concentration varies. The critical pressure p_c above which the ferroelectric VI phase appears is shown in Fig. 11 as a function of x. The experimental point corresponding to $p=0$ is determined from the x-T phase diagram at atmospheric pressure measured previously.[4] The critical pressure p_c varies nearly linearly with the deuterium concentration x in the region $x \lesssim 0.5$. As the deuterium concentration further increases a deviation from the linear relation becomes progressively large. The

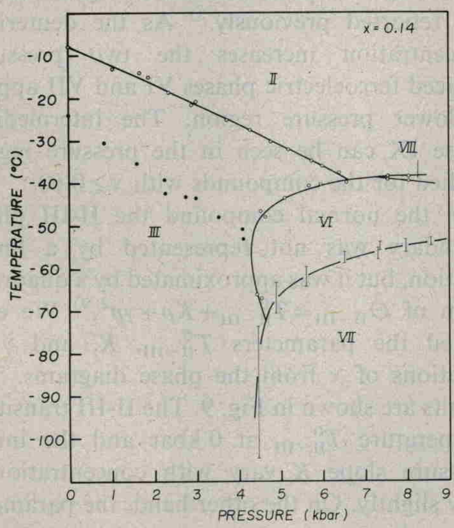

Fig. 4. Pressure-temperature phase diagram of $\{(NH_4)_3H(SO_4)_2\}_{0.86}\{(ND_4)_3D(SO_4)_2\}_{0.14}$. Solid circles and vertical bars: see the caption of Fig. 3.

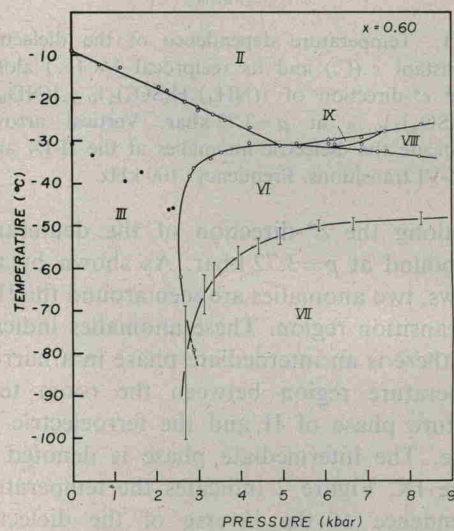

Fig. 6. Pressure-temperature phase diagram of $\{(NH_4)_3H(SO_4)_2\}_{0.40}\{(ND_4)_3D(SO_4)_2\}_{0.60}$. Solid circles and vertical bars: see the caption of Fig. 3.

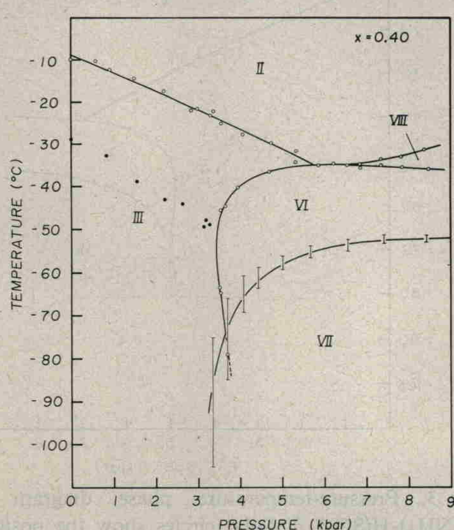

Fig. 5. Pressure-temperature phase diagram of $\{(NH_4)_3H(SO_4)_2\}_{0.60}\{(ND_4)_3D(SO_4)_2\}_{0.40}$. Solid circles and vertical bars: see the caption of Fig. 3.

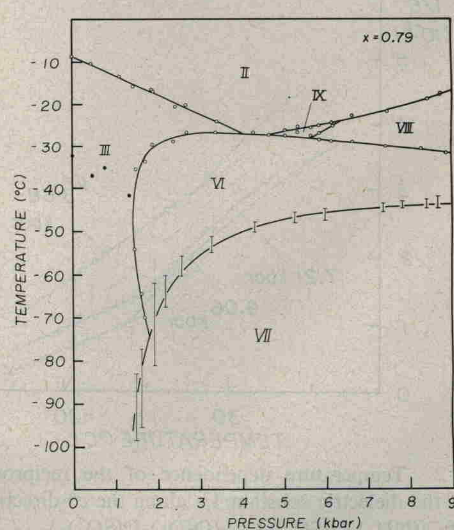

Fig. 7. Pressure-temperature phase diagram of $\{(NH_4)_3H(SO_4)_2\}_{.021}\{(ND_4)_3D(SO_4)_2\}_{0.79}$. Solid circles and vertical bars: see the caption of Fig. 3.